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Investigation of the Statistical Properties of Light

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Abstract

The report describes the coherence properties and the statistical properties of light from a classical and quantum mechanical point of view. The theoretical part is used to describe some more specific examples within the field of light scattering such as light scattering from a collection of independent scatters, measurement of wind velocity, and single burst detection in a Laser-Doppler velocitimeter system where the description gives a figure of merit for such a system. An experimental investigation was made of Brillouin scattering in some organic liquids. The experimental equipment is described and the results reported.

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1. INTRODUCTION

This report consists of two parts; the theoretical part concerns the concept of light and gives a description of the statistical properties of light in terms of correlation functions. The second part describes some examples within the field of light scattering.

In the first part it is intended to give a general description of the statistical features of light and the concept of optical coherence.

The statistical features of light are expressed by the way light is generated. Thermal light is generated by many uncorrelated radiators (atoms) and laser light is generated through a collective interaction between the electromagnetic field and a collection of atoms. The statistical properties could also result from a stochastic process; i.e. light scattering by a collection of scattering centres which in themselves constitute a stochastic dynamical system.

Now, in many cases we cannot calculate the probability distributions for the light field in an explicit way even though we have full information about the light source, and often we are not interested in the full amount of information but only in the first few moments of the distribution functions for the field. This leads to a description of the properties of light by means of a hierarchy of correlation functions, or to put it in another way, a description of the coherence properties of the electromagnetic field. The description will be complete if we know the correlations to any order.

Historically, the concept of optical coherence is connected to the interference of light. Light fields were said to be coherent if the field in two space-time points was able to interfere, visualized as a fringed pattern in space or time, and said to be incoherent if no such pattern was observable. Such a description is an ideal one, for real optical fields the visibility of the interference patterns will differ for different beams and for different space-time-points, so a more quantitative description must be introduced through the concept "partial coherence" as an intermediate state of the coherence and incoherence states. Furthermore, we must specify the "order of coherence"; in an experiment where we observe interference between the amplitudes of two fields we talk about "second-order coherence", and when we observe interference between the intensities of two beams we investigate the "fourth-order coherence" properties of the fields. Later on a more general definition of the order of coherence is given.

Earlier theoretical and experimental investigations of light beams primarily concerned the second-order coherence properties of the fields; some of the most important works here are Verdet (1859)[1], von Laue (1907)[2], Michelson (1890), Van Cittert (1934)[3], Zernike (1938)[4] and Mandel and Wolf (1961)[5]. In 1956 Hanbury Brown and Twiss[6] opened up a new field of optics with their famous intensity-correlation experiment. The experiment and the interpretation of the results show the necessity of developing a more general statistic description of light phenomena. Such a theory can be developed from two points of view, one classical, based on the wave concept of light, and one quantum-mechanical, based on the partial (photon) concept of light. In 1962 Wolf[7] proposed a classical theory in which all orders of coherence were included and in 1963 Glauber[8] developed a fully quantum-mechanical theory of the coherence of light.

To gain information about the light we must make a measurement, and in this respect the theory of coherence has the property of only dealing with measurable quantities. In the classical Maxwell theory of electromagnetic fields, the electric and magnetic fields were, in principle, measurable quantities, and when these fields could not be measured at optical frequencies, it was due to the fact that no real detector could follow the oscillation of the field. Classically, the measurable quantities of light are the intensity averaged over a cycle of oscillations. In quantum language this is expressed by saying that a light detector is a "quantum detector". A quantum detector is based on the photoelectric process in which the energy of a light beam can only be removed in light quanta (photons), or we could say that in detecting light we must destroy it (another way to detect light would be to create photons). As a consequence, the measured intensity is proportional to the product of the positive and negative frequency part of the electrical field. In this text we will follow the quantum point of view with respect to the detection process.

Part two deals with light scattering from different set-ups. Chapter 6 treats the measurement of wind velocity in the atmosphere, the main problem being the general one, the propagation of a light beam through an inhomogeneous medium. In the case of strong disturbances, i.e. a high turbulence level, the problem is hard to solve because of the effect of multi-scattering on the medium, and to get a reasonable description in mathematical terms, one must restrict oneself to only consider small disturbances in the scattering medium. In the proposed set-ups for measuring the cross-wind velocity, the effect of the disturbances in different areas along the light beam paths is investigated to describe the information obtainable from such measurements.

Chapter 7 deals with the detection of low-level signals from single particles in a Doppler-velocitimeter system. The signals are stochastic, transient signals because of the photo detection process. In the treatment of the problem it appears that one can give a figure of merit for the Doppler system that can be used to adjust the system parameters to obtain the greatest amount of information in actual measurements with the given set-ups.

In chapter 8 the main problem considered is whether, in the case of light scattering from a collection of independent scattering centres, the electric field has Gaussian statistics or not. The scattering of light from particles undergoing Brownian motion and from particles suspended in a turbulent flow is investigated.

The last chapter reports on experimental equipment used to investigate Brillouin scattering in liquids. The results from the experiment are used for comparison with results predicted by the Landau-Placzek relaxation theory and by the Mandel'shtam and Leontovich relaxation theory. It appears that the Mandel'shtam-Leontovich theory best fits the results obtained in this work for some organic liauids.

2. CLASSICAL DESCRIPTION OF THE COHERENCE OF LIGHT

2.1. Complex Representation of Real Fields

In the classical Maxwell theory of electromagnetic fields, the electric and magnetic fields are represented by real quantities, and it could seem artificial to introduce complex field quantities. But when we deal with light and light detectors, the complex field has a direct physical meaning, as we shall see later on. Further, it serves as a bridge between a classical and a quantum description of light. As mentioned in the introduction, the detection process of light described in a purely classical way is a cycle averaging, i.e. the intensity of the incoming light is proportional to the square of the real electric field, and the output of the light detector is these quantities averaged over a cycle of oscillations. The loss of information about the phase of the electric field in the detection process is in this case due to the time of resolution for the detector. Now, this concept is incorrect, even though it gives the correct result; a light detector is based on the photoelectric effect (2.1), which can only be described in a quantum-mechanical way, and, according to this, the energy of the light beam is removed from the beam in light quanta, photons, the photodetector destroys the incoming photons. The quantum-mechanical operator for photon annihilation can be shown to be the positive frequency part of the real electric field and the measured intensity will be proportional to the

absolute square of this quantity. Thus, if we wish to give a description of light in classical terms, which is consistent with a quantum-mechanical description, use of a complex notation is suitable.

If we consider a real quantity $V^{r}(t)$, which could be a scalar or a vector quantity related to the electromagnetic field, and assume it has a Fourier transformed, we can write the Fourier transformed of $V^{(r)}(t)$ as:

$$V(\omega) = \int_{-\infty}^{\infty} V^{(r)}(t)\, e^{-i\omega t}\, dt\,, \tag{2.1.1}$$

and define

$$V^{+}(\omega) = \begin{cases} V(\omega) & \omega \geq 0 \\ 0 & \omega < 0 \end{cases} \tag{2.1.2}$$

and

$$V^{-}(\omega) = \begin{cases} 0 & \omega > 0 \\ V(\omega) & \omega \leq 0 \end{cases} \tag{2.1.3}$$

From $V^{+}(\omega)$ and $V^{-}(\omega)$, we define the complex field quantities in the time domain as:

$$V^{+}(t) = \frac{1}{\pi} \int_{-\infty}^{\infty} V^{+}(\omega)\, e^{i\omega t} dtv \tag{2.1.4}$$

and

$$V^{-}(t) = \frac{1}{\pi} \int_{-\infty}^{\infty} V^{-}(\omega)\, e^{i\omega t} dtv\,. \tag{2.1.5}$$

From these equations we get:

$$V^{(r)}(t) = \frac{1}{2}(V^{+}(t) + V^{-}(t))$$

and

$$V^{(i)}(t) = \frac{1}{2i}(V^{+}(t) - V^{-}(t))\,, \tag{2.1.6}$$

where $V^{(i)}$ is the imaginary part of the complex field. $V^{(i)}$ contains the same physical information as $V^{(r)}(t)$, as we shall see. A condition for the existence of a Fourier transform is ordinarily stated to be that the actual function must be absolutely integrable. This is a very restrictive demand,

and it excludes a broad class of function, for instance, the Dirac delta function. To overcome this, we may demand that the Fourier transform exist in a Chauchy way:

$$f(\omega) = \lim_{\varepsilon \to o} \int_{-\infty}^{\infty} f(t)\, e^{-i\omega t} \cdot e^{-\varepsilon|t|} dt \qquad \varepsilon > o \qquad (2.1.7)$$

or, when we transfer from frequency space to time space,

$$g(t) = \lim_{\varepsilon \to o} \frac{1}{2\pi} \int_{-\infty}^{\infty} g(\omega)\, e^{i\omega t} \cdot e^{-\varepsilon|\omega|} d\omega \qquad \varepsilon > o \qquad (2.1.8)$$

For instance, the Dirac delta function can be expressed as:

$$\delta(t) = \lim_{\varepsilon \to o} \frac{1}{2\pi} \int_{-\infty}^{\infty} 1 \cdot e^{i\omega t} \cdot e^{-\varepsilon|\omega|} d\omega = \lim_{\varepsilon \to o} \frac{1}{\pi} \frac{\varepsilon}{\varepsilon^2 + t^2} \qquad (2.1.9)$$

and for the function

$$\eta(\omega) = \begin{cases} 1 & \omega \geq o \\ o & \omega < o \end{cases} \qquad (2.1.10)$$

we get:

$$\eta(t) = \lim_{\varepsilon \to o} \frac{1}{2\pi} \int_{-\infty}^{\infty} \eta(\omega)\, e^{i\omega t} e^{-\varepsilon|\omega|} d\omega = \lim_{\varepsilon \to o} \frac{1}{2\pi} \frac{1}{\varepsilon - it}$$

$$= \frac{1}{2}\delta(t) - \lim_{\varepsilon \to o} \frac{1}{2\pi i} \frac{t}{\varepsilon^2 + t^2} = \frac{1}{2}\left(\delta(t) - \frac{1}{\pi i} P \frac{1}{t}\right) \qquad (2.1.11)$$

where P is a symbolic expression for the above limit, and it denotes the principal value of a given function.

Now, by using this technique, the complex fields can be expressed as:

$$V^{+}(t) = \frac{1}{\pi}\int_{-\infty}^{\infty} V^{+}(\omega)e^{i\omega t}dt$$

$$= \frac{1}{\pi}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} V^{(r)}(t')\eta(\omega)e^{i\omega t}e^{-i\omega t'}\,dt\,dt'$$

$$= \int_{-\infty}^{\infty} V^{(r)}(t')\left(\delta(t-t') - \frac{1}{\pi i}P\frac{1}{t-t'}\right)dt' \qquad (2.1.12)$$

$$= V^{(r)}(t) - \frac{1}{\pi i}P\int_{-\infty}^{\infty}\frac{V^{(r)}(t')}{t-t'}\,dt'$$

and we obtain the relations

$$V^{(i)}(t) = \frac{1}{\pi}P\int_{-\infty}^{\infty}\frac{V^{(r)}(t')}{t-t'}\,dt', \qquad (2.1.13)$$

which means that the imaginary field is the Hilbert transform of the real field, and as stated, no new information is contained in $V^{(i)}(t)$, and the full amount of physical information about the field is contained in the complex field as well as in the real field. The complex field will further obey the Maxwell equations of the field, a property which can be easily proved by insertion of the complex field in the Maxwell equation.

2.2. Correlation Functions and the Statistical Description of Light

In order to define the field correlation functions we first give a stochastic description of light. The description is based on the classical theory of stochastic processes, and the definition given here can be used in other connections where we deal with stochastic processes.

In a classical theory of stochastic processes, the statistical properties of a set of quantities are described by a probability density which is restricted to be positive and smaller than one. Quantum mechanically, one can show a formal equivalence between the classical and the quantum description by introducing the so-called Glauber-Sudarshan representation[9,10], although the equivalent "probability density" in the quantum description is not restricted to positive values.

If we denote the complex field in a space-time point (x_j, t_j) as:

$$V_j^+ = V^+(x_j, t_j),$$

we can describe the statistical properties of the field quantities by a sequence of probability density functions:

$$P_1(V_1^+ \; V_1^-),\; P_2(V_1^+ \; V_1^- \; V_2^+ V_2^-) \;\ldots\; P_n(V_1^+ \; V_1^- \; \ldots \; V_n^+ V_n^-) \ldots \quad (2.2.1)$$

where P_n, the n-fold joint probability density, can be interpreted by the expression,

$$P_n(V_1^+ V_1^- \; \ldots \; V_n^+ V_n^-) dV_1^+ \, dV_1^- \; \ldots\ldots \; dV_n^+ \, dV_n^- \,, \qquad (2.2.2)$$

which gives the probability that

$$V^+(x_1 t_1) \quad \text{and} \quad V(x_1 t_1)$$

lie in an interval $dV_1^+ \, dV_1^-$ about V_1^+ and V_1^-

and

$$V^+(x_2 \; t_2) \quad \text{and} \quad V^-(x_2 \; t_2)$$

lie in an interval $dV_2^+ dV_2^-)$, about V_2^+ and V_2^-, and

$$\vdots$$

$$V^+(x_n \; t_n) \quad \text{and} \quad V^-(x_n \; t_n)$$

lie in an interval $dV_n^+ \, dV_n^-$ about V_n^+ and V_n^-.

In general, V_j^+ is a spatial vector and the joint n-fold probability density is a function with 6n variables.

Now, given a function F which depends on the field in n space-time points, we can define the ensemble average of F by the relation:

$$\langle F(V_1^+ V_1^- \; \ldots \; V_n^+ V_n^-) \rangle = \int F \cdot P_n \, dV_1^+ \; \ldots \; dV_n^+ dV_n^- \,. \qquad (2.2.3)$$

The physical meaning of an ensemble averaging is the mean of a quantity measured from an infinite series of experiments with different samples of the same system. The description of a physical system by ensemble-averaged quantities is, in general, the only way to obtain a meaningful description; the time-averaged quantities which are ordinarily measured in the process can, if the system is stationary and ergodic, be related to the ensemble average by the relation

$$\langle F \rangle = \lim_{T \to \infty} \frac{1}{T} \int_{t}^{t+T} f \, dt \, .$$

But in general the time-averaged quantity itself will be a stochastic variable, whether the system is stationary or not.

Now, with this in mind, we define the correlation functions or more precisely the correlation tensor of field as:

$$\Gamma^{(n,m)}(x_1 t_1 \; x_2 t_2 \; \ldots \; x_{n+m} \; t_{n+m}) = \langle V^+(x_1 t_1) \ldots V^+(x_n \; t_n) \, V^-(x_{n+1} \; t_{n+1}) \ldots V^-(x_{n+m} \; t_{n+m}) \rangle \tag{2.2.4}$$

The order of the correlation function is defined by $n+m$. Knowledge of the entire correlation tensor gives us full information about the field. From the component of the correlation tensor we can construct the characteristic function of the field, which is defined as the Fourier transform of the probability density and which acts as generating function for the correlation functions. By Fourier transforming the characteristic function we obtain the probability density.

For the Fourier transform of the fields we can further define a spectral correlation function $G^{(n,m)}$

$$G^{(n,m)}(x_1 \omega_1 \; x_2 \omega_2 \; \ldots \; x_{n+m} \; \omega_{n+m}) = \left(\frac{1}{2\pi}\right)^{n+m} \langle V^+(x_1 \omega_1) \ldots V^+(x_n \; \omega_n) \, V^-(x_{n+1} \; \omega_{n+1}) \ldots V^-(x_{n+m} \; \omega_{n+m}) \rangle \tag{2.2.5}$$

where

$$V^+(x_1 \omega_1) = \int_{-\infty}^{\infty} V^+(x_1 t_1) \, e^{-i\omega_1 t_1} \, dt_1 \, . \tag{2.2.6}$$

By inserting this expression in eq. 2.1, we obtain

$$G^{(n,m)}(x_1\omega_1 x_2\omega_2 \cdots x_{n+m}\ \omega_{n+m}) =$$

$$(\tfrac{1}{2\pi})^{n+m} \int \Gamma^{(n,m)}(x_1 t_1 \cdots x_{n+m} t_{n+n}) e^{-i\omega_1 t_1 \cdots i\omega_{n+m} t_{n+n}}$$

$$dt_1 dt_2 \cdots dt_{m+m} . \qquad (2.3.7)$$

In the case of stationary fields, we can obtain some important relations for the correlation functions. By using eq. 2.2.4, we obtain for the second-order correlation function:

$$\Gamma^{(n,m)}(x_1 t_1 \quad x_2 t_1 + \tau) = \Gamma^{(1,1)}(x_1 x_2, \tau) = \delta_{n,m} \langle V^+(x_1 t_1) V^-(x_1 t_1 + \tau) \rangle \qquad (2.2.8)$$

which means that only the positive and negative frequency parts are correlated to second order. For the spectral correlation function, we obtain:

$$G^{(1,1)}(x_1 \omega_1 x_2 \ \omega_2) = \frac{1}{2\pi} \delta(\omega_1 + \omega_2) \int \Gamma^{(1,1)}(x_1 x_2, \tau) e^{-i\omega_2 \tau} d\tau$$

$$= \delta(\omega_1 + \omega_2)\, S^{(1,1)}(x_1 x_2, \omega_2) , \qquad (2.2.9)$$

where $S^{(1,1)}(x_1 x_2, \omega_2)$ can be interpreted as the spectral density of the fields. The relation between the spectral density and the second-order correlation function is known as the Wiener-Khintchine theorem

$$S^{(1,1)}(x_1 x_2, \omega) = \frac{1}{2\pi} \int \Gamma^{(1,1)}(x_1 x_2, \tau)\, e^{-i\,\omega\tau} d\tau . \qquad (2.2.10)$$

The result obtained in eq. 2.2.8 can be extended to be valid for all orders in some special but important cases. First, if the correlation function is stationary in both time and space (invariant under a spatial translation), it can be shown (10) that

$$\Gamma^{(n,m)}(x_1 t_1 \cdots x_{n+m}, t_{n,m}) =$$

$$\delta_{n,m} \langle V^+(x_1 t_1) \cdots V^+(x_n\ t_n) \cdot V^-(x_{n+1}\ t_{n+1}) \cdots V^-(x_{2n}\ t_{2n}) \rangle , \qquad (2.2.11)$$

A second case is when the spectral quantities of the field

$$V^{+}(x_1,\omega) = \int_{-\infty}^{\infty} V^{+}(x_1,t)\, e^{-i\omega t} dt$$

only differ from zero in a small range about a centre frequency ω_o. In this case $\Gamma^{(n,m)}$ will only differ from zero when V^{+} and V^{-} act in pairs. To understand this, we consider the spectral correlation function:

$$G^{(n,m)}(x_1\omega_1 \ldots\ldots x_{n+m}\ \omega_{n+m})$$

$$= (\tfrac{1}{2\pi})^{n+m} \langle V^{+}(x_1\omega_1) \ldots V^{+}(x_n\ \omega_n) V^{-}(x_{n+1}\ \omega_{n+1}) \ldots V^{-}(x_{n+m}\ \omega_{n+m}) \rangle$$

$$= \delta(\Sigma_{p=1}^{n+m} \omega_p)\ S^{(n,m)}(x_1 \cdots x_{n+m},\ \bar{\omega}) \qquad (2.1.12)$$

where $S^{(n,m)}$ is the mutual spectral density that is related to the correlation function $\Gamma^{(n,m)}$ through the generalized Wiener-Khintchine theorem

$$S^{(n,m)}(x_1 \cdots x_{n+m},\bar{\omega}) = (\tfrac{1}{2\pi})^{n+m-1} \int \Gamma^{(n,m)}(x_1 x_2 \cdots x_{n+m};\bar{\tau}) e^{-i\bar{\omega}\cdot\bar{\tau}}\ d\bar{\tau}$$

where

$$\bar{\omega} = (\omega_2, \ldots\ldots, \omega_{n+m})$$

and

$$\bar{\tau} = (t_2 - t_1, \ldots\ t_{n+m} - t_1). \qquad (2.1.13)$$

From eq. 2.2.12 we obtain the condition:

$$\sum_{p}^{n+m} \omega_p = 0,$$

which can only be fulfilled for $n \neq m$ when at least one frequency, say ω_{m+1}, is far from its centre frequency $-\omega_o$. This means that $G^{(n,m)}$ is zero according to the above assumption for $V^+(x, \omega)$, and this further implies that $\Gamma^{(n,m)}$ is zero (or practically zero) for $n \neq m$. So, if we can consider the spectral widths of field quantities to be small compared with the centre frequency, then eq. 2.2.11 is valid.

2.3. The Degree of Coherence of Light

The correlation function is an important tool in describing the coherence properties of light. The correlation function expresses the amount of "noise" in the field quantities, and by investigating the correlation functions to a certain degree of order we can compensate for our lack of full knowledge about the light. A correlation theory, as stated here, has many similarities in other branches of physics, for example, the improvement of the theory of turbulance in fluid dynamics. It is perhaps astonishing that the development of a complete theory of the coherence of light, classical and quantum-mechanical, is of relatively new date. An explanation of this fact must be that the light sources available before 1960 were of a chaotic nature, and in describing the properties of these cources it was sufficient to investigate the second-order properties. First with the appearance of the laser was the need for a more complete theory obvious.

Now, in order to define a measure for the coherence properties of light, we must first specify the fully coherent state, and we will define it as a noiseless state, or the state where the field quantities are deterministic quantities. This means that the probability density for the field quantities must be a product of δ-functions, and with it the correlation function must be a product of the coherent field quantities, so we can write:

$$\Gamma_c^{(n+m)}(x_1 t_1 \ldots x_{m+n}) = V_c^+(x_1 t_2) \ldots V_c^+(x_n t_n)\, V_c^-(x_{n+1} t_{n+1}) \ldots V_c^-(x_{n+m} t_{n+m}) \tag{2.3.1}$$

To define the degree of coherence, we demand that the absolute value of this quantity is one in the fully coherent case, and we will use a definition given by Glauber[8], though other definitions are available[11,12]. The degree of coherence $\gamma^{(n,m)}$ to the order 2n is then expressed by:

$$\gamma^{(n,n)}(x_1t_1 \cdots x_{2n}t_{2n}) = \frac{\Gamma^{(n,n)}(x_1t_1 \cdots\cdots x_{2n}t_{2n})}{[\Gamma^{(1,1)}(x_1x_1t_1)\Gamma^{(1,1)}(x_2x_2t_1)\cdots\cdots\Gamma^{(1,1)}(x_{2n}x_{2n}t_{2n})]^{\frac{1}{2}}} \qquad (2.3.2)$$

and we see that the absolute value of $\gamma^{(n,n)}$ is one in the fully coherent case by using eq. 2.3.1. It can be shown (2.6) that if we define the full coherence to the order of 2n by:

$$|\gamma^{n,n}| = 1, \qquad (2.3.3)$$

this will imply a factorization of $\Gamma^{(n,n)}$, as expressed in eq. 2.3.1.

The factorization properties of the correlation functions in the case of full coherence suggest a mode description for partially coherent light, so a general correlation function can be expressed as a sum of coherent contributions.

In the following section this mode description is used in the case of stationary fields to investigate the second-order coherence properties.

3. SECOND-ORDER COHERENCE

3.1.

The second-order coherence properties of light fields are described in all textbooks dealing with the coherence of light, and classical examples illustrating the ideas of temperal and spatial coherence are, respectively, the Michelson interferometer and the Young interference experiments. The ability of a light beam to show interference patterns is closely related to its second-order coherence properties, and in fact a light beam was said to be coherent, i.e. by similar to a classical stable wave,in the areas where interference could occur, before the famous Hanbury Brown and Twiss experiment where it was shown that light, which was coherent to second order, was completely incoherent in fourth order.

This part seeks to describe the second-order coherence properties in a mode description which has a connexion with the quantum mechanical concept of the photon. A photon is a "particle" which has a momentum and an energy; the energy is related to a frequency and the momemtum to a frequency and a direction. The mode number describes, as we shall see, the same features. The idea in a mode description is to express the second-order correlation function as a sum of terms that each are coherent to second order. To do this we will express $\Gamma^{(1,1)}$ by the spectral density (eq. 2.2.10)

$$\Gamma^{(1,1)}(x_1 x_2, \tau) = \int S^{(1,2)}(x_1 x_2, \omega)\, e^{i\omega\tau} d\omega \tag{3.1.1}$$

and then attempt to express $S^{(1,1)}(x_1\ x_2, \omega)$ as:

$$S^{(1,1)}(x_1 x_2, \omega) = \sum_{\Delta} a(\Delta, \omega)\ U^{*}_{\Delta,\omega}(x_1) U_{\Delta,\omega}(x_2), \tag{3.1.2}$$

where the mode functions $U_{\Delta,\omega}$ must satisfy the same wave equation as the field quantities, and further it must form a complete orthonormal set of functions which satisfy the integral equation:

$$\int S^{(1,1)}(x_1 x_2, \omega) U_{\Delta,\omega}(x_2) dx_2 = a(\Delta, \omega) U_{\Delta,\omega}(x_1). \tag{3.1.3}$$

The eigenvalue $a(\Delta, \omega)$ determines the strength by means of which the mode (Δ, ω) is excited. The eigenvalues will be real and positive quantities, a fact which is due to the Hermitian properties of $S^{(1,1)}$, i.e.:

$$\left(S^{(1,1)}(x_1 x_2, \omega)\right)^{*} = S^{(1,1)}(x_2\ x_1, \omega). \tag{3.1.4}$$

The eigenvalue can be expressed as:

$$a(\Delta, \omega) = \int S^{(1,1)}(x_1 x_2, \omega) U_{\Delta,\omega}(x_2) U^{*}_{\Delta,\omega}(x_1) dx_1 dx_2 \tag{3.1.5}$$

and it can be interpreted as the energy density in the mode (Δ, ω), and by using the completeness of the functions $U_{\Delta,\omega}$ we obtain the relations:

$$\sum_{\Delta} a(\Delta, \omega) = \int S^{(1,1)}(x_1\ x_1, \omega) dx_1, \tag{3.1.6}$$

which is the energy density at the frequency ω in the space under consideration.

Now, we can write the second-order correlation functions in the desired forms as a sum of fully coherent terms

$$\Gamma^{(1,1)}(x_1 x_2, \tau) = \int \sum_{\Delta} a(\Delta,\omega)\, U^{*}_{\Delta,\omega}(x_2) U_{\Delta,\omega}(x_2)\, e^{i\omega\tau}\, d\omega \qquad (3.1.7)$$

or, if we numerate the frequencies, we obtain

$$\Gamma^{(1,1)}(x_1 x_2, \tau) = \sum_{\Delta,n} a(\Delta,n)\delta\omega\, U^{*}_{\Delta,n}(x_1)\, U_{\Delta,n}(x_2) e^{i\omega_n \tau}\ , \qquad (3.1.8)$$

where $\Delta\omega$ is a small frequency increment.

From eq. 3.1.8 we see that if we can perform an optical filtering process, i.e. exclude all modes except one, we can create a light beam from an arbitrary source that is coherent to second order.

To obtain a more concrete picture of the features of eq. (3.1.7), we will consider a special, but important case where the spectral density is stationary in space, i.e.

$$S^{(1,1)}(x_1 x_2 \omega) = S^{(1,1)}(x_2 - x_1, \omega)\ . \qquad (3.1.9)$$

Further, by assuming a finite world with periodical boundaries, which is tantamount to restricting ourselves to only consider this finite world in an experimental situation, we see that the eigenfunctions to the integral equation (3.1.3) are plane waves;

$$U_{\bar{k},\omega}(\bar{r})\ \frac{1}{V^{1/2}}\ e^{i\bar{k}\cdot\bar{r}}\ ,$$

where V is the volume of our world and $\bar{k}$ the wave vector which must fulfil the condition;

$$|\bar{k}| = \omega/c\ ,$$

where c is the velocity of light.

Now, we can write eq. (3.1.7) as:

$$\Gamma^{(1,1)}(r_1 r_2, \tau) = \frac{1}{V} \int\sum a(\bar{k},\omega) 2^{-i\bar{k}\cdot(r_2 - r_1)} \cdot e^{i\omega\tau}\, d\omega\ , \qquad (3.1.10)$$

where the summation is extended over all directions of the wave vector $\bar{k}$

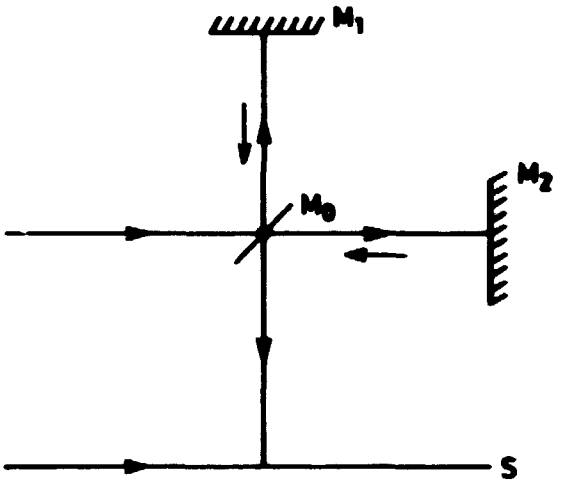


Fig. 3.1

3.2. Temporal coherence

In the case of the Michelson interferometer (see fig. 3.1), the incoming light beam is pre-filtered so only one directional mode is present. The field at the screen is a superposition of the field from the two branches of the interferometer;

$$V^+(t) = V_1^+(t) + V_2^+(t) \qquad (3.2.1)$$

where

$$V_1^+(t) = a_1 V_o^+(t)$$

and

$$V_2^+(t) = a_2 V_o^+(t+\tau)$$

where V_o^+ is the incoming field, a_1 and a_2 are constants depending on the transmission and reflection properties of the mirrors, and τ is the time delay due to the different path lengths in the two branches of the interferometer

$$\tau = L/c .$$

The mean intensity at the screen then becomes:

$$I_s = \Gamma^{(1,1)}(x_1 t_1 x_1 t_1) = |a_1|^2 \Gamma_o^{(1,1)}(x_1 x_1) + |a_2|^2 \Gamma_o^{(1,1)}(x_1 x_1) + a_1 a_2^* \Gamma_o^{(1,1)}(x_1 x_1 \tau) + a_1^* a_2 \Gamma_o^{(1,1)}(x_1 x_1, -\tau) \tag{3.2.2}$$

where

$$\Gamma_o^{(1,1)}(x_1 x_1, \tau) = \langle V_o^+(x_1 t) V_o^-(x_1 t+\tau) \rangle .$$

From eq. (3.1.10) we have

$$\Gamma_o^{(1,1)}(x_1 x_1 \tau) = \frac{1}{V} \int a(\omega) e^{i\omega\tau} d\omega = \frac{1}{V} e^{i\omega_o \tau} \int a(\omega+\omega_o) e^{i\omega\tau} d\omega = e^{i\omega_o \tau} g(\tau) \tag{3.2.3}$$

where ω_o is the centre frequency of $a(\omega)$. In the case of a symmetrical spectrum, $g(\tau)$ will be a non-oscillating and real function of τ; this will be assumed here, and for the intensity we obtain:

$$I_S \propto 1 + \frac{2|a_1||a_2|}{|a_1|^2 + |a_2|^2} \frac{g(\tau)}{g(0)} \cos(\omega_o \tau + \alpha) . \tag{3.2.4}$$

We see that interference will only occur when τ is smaller than a characteristic decay time τ_c of $g(\tau)$, the coherence time of the light beam.

In general, τ_c is related to the spectral width of $a(\omega)$ by

$$\tau_c \Delta\omega = 1 ,$$

and the condition of interference or coherence of the light at the screen can be stated as

$$\tau \Delta\omega < 1 \tag{3.2.5}$$

or

$$L < c/\Delta\omega = \Lambda_c , \tag{3.2.6}$$

where Λ_c can be interpreted as a coherence length of the light.

The condition stated in eq. 3.2.5 of coherence to second order is a weakening of the definition of full coherence stated in eq. 2.3.3, according to which coherence is only achieved for a τ value that fulfils the condition;

$$g(\tau)/g(0) = 1,$$

but the condition in eq. 3.2.5 is more practical, and must be interpreted as "near" coherence of an actual light beam.

3.3. Young's experiment

The Young experiment can be treated in almost the same way as the Michelson interferometer experiment. An experiment of the Young type is sketched in fig. 3.2; the lens acts as an optical filter which only allows the presence of modes in the z-direction. Plane A is provided with two

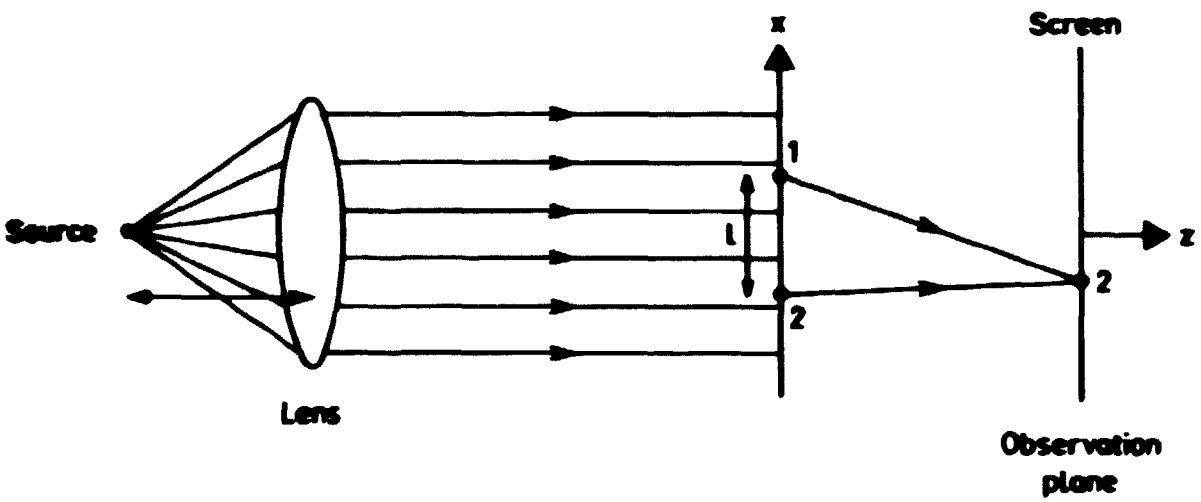


Fig. 3.2

pinholes separated a distance l in the x-direction. The light propagating from the two pinholes hits the observation plane, and we find the condition for obtaining an interference picture on this screen. Using the designation of fig. 3.2, the field at a point Q in the observation plane is

$$V_Q^+(t) = a_1 V^+(x_1, t+r_1/c) + a_2 V^+(x_2, t+r_2/c), \qquad (3.3.1)$$

where a_1 and a_2 are constants, depending on the size and shape of the pinhole and the distance to the point Q. V^+ is the filtered field from the source. Now, in the same way as in the Michelson experiment, we obtain, for the intensity at the point Q, by using eq. 3.1.10

$$I_Q \propto 1 + \frac{2|a_1||a_2|}{|a_1|^2+|a_2|^2} \frac{g(\tau + \frac{x_1-x_2}{c})}{g(o)} \cos\omega_o(\tau + \frac{x_2-x_1}{c} + \alpha_{12}) \quad (3.3.2)$$

where

$$\tau = r_1/c - r_2/c$$

and α_{12} is a phase factor due to the constants a_1 and a_2. From eq. 3.3.2, we see that fringes with maximum intensity are formed when

$$\omega_o(\tau + \frac{x_2-x_1}{c} + \alpha_{12}) = p\pi ,$$

where p is an integer.

The condition of interference or coherence is

$$|\tau + \frac{x_1-x_2}{c}| \ll 1, \quad (3.3.3)$$

which is equivalent to the condition obtained in the Michelson experiment.

3.4. Coherence to fourth order

Finally, we here consider some cases involving fourth-order coherence properties of light, which can be described in terms of the second-order correlations function. If the field quantities have a Gaussian distribution, the fourth-order correlation function can be written in terms of the second-order function by means of the Siegert relation (see Appendix A).

$$\Gamma^{(2,2)}(x_1t_1x_2t_2\ x_3\ t_3\ x_4t_4) = \Gamma^{(1,1)}(x_1t_1x_3t_3)\Gamma^{(1,1)}(x_2t_2\ x_4t_4) \quad (3.4.1)$$

$$+ \Gamma^{(1,1)}(x_1t_1x_4t_4)\Gamma^{(1,1)}(x_2t_2\ x_3t_3) .$$

The first example is equivalent to the Hanbury Brown and Twiss experiment. Let us assume that an incoming stationary light beam is prefiltered so that only one mode is present in the beam. The light beam is then split into two parts, and the intensities of the two beams are detected by two distinct photodetectors placed at the space points, x_1 and x_2. By correlating the two intensities, we can measure the fourth-order correlation function:

$$\langle I_1(t) I_2(t) \rangle \propto \Gamma^{(2,2)}(x_1 t_1 x_2 t_2 \; x_1 t_1 x_2 t_2)$$

$$= \Gamma^{(1,1)}(x_1 x_1, 0)\, \Gamma^{(1,1)}(x_2 x_2, 0) + |\Gamma^{(1,1)}(x_1 x_2, \tau)|^2 \qquad (3.4.2)$$

$$= \Gamma^{(1,1)}(x_1 x_1, 0)\, \Gamma^{(1,1)}(x_2 x_2, 0) \; [1 + |\gamma^{(1,1)}(x_1 x_2, \tau)|^2]$$

where τ is a measure of the different path lengths of the light at the two detectors. From eq. 3.4.2 we obtain:

$$\gamma^{(2,2)}(x_1 x_2, \tau) = 1 + |\gamma^{(1,1)}(x_1 x_2, \tau)|^2 \qquad (3.4.3)$$

and we see that in the case where only one mode is present in the light beam, i.e. the light is coherent to second order, the light will be completely incoherent to fourth order. Further, if the light beam is fully incoherent to second order, it will be coherent to fourth order in the space-time point $(x_1 x_2, \tau)$. This emphasizes that care must be taken when using the word "coherence", and one must always specify the order of coherence under consideration. Equation 3.4.3 can, in quantum mechanical terms, be interpreted as a bunching effect of photons originating from a chaotic source, or as a tendency of the photons to arrive in bundles.

In the foregoing examples, we have implicitly assumed that the detector was a point detector, and have not taken into account the effect of the detector area. The photo-detector current is proportional to the sum of the light intensities at the space point belonging to the detector surface, i.e.

$$i(t) \propto \int_A I(x_1 t)\, dA, \qquad (3.4.4)$$

where the integration takes place over the surface A of the photodetector.

From eq. 3.4.4 we can obtain the autocorrelation function of the current;

$$\langle i(t)\, (i(t+\tau)) \rangle = \int_A \int_A \langle I(x_1 t)\, I(x_2 \; t+\tau) \rangle \, dA_1 dA_2 \qquad (3.4.5)$$

$$= \int_A \int_A \Gamma^{(2,2)}(x_1 t_1 x_2 \; t+\tau \; x_1 t_1 x_2 \; t+\tau)\, dA_1 dA_2$$

and we see that by measuring the autocorrelation functions of the current, we are measuring the fourth-order correlation functions integrated over the detector area. In the case of Gaussian distribution of the field quantities, we obtain by using eq. 3.4.1

$$\begin{aligned} \langle i(t)i(t+\tau)\rangle &= \int_A\int_A \Gamma^{(1,1)}(x_1x_1o)\Gamma^{(1,1)}(x_2,x_2,o)dA_1dA_2 \\ &+ \int_A\int_A |\Gamma^{(1,1)}(x_1x_2,\tau)|^2 dA_1dA_2 \qquad (3.4.6) \\ &= \langle i\rangle^2 + \int_A\int_A |\Gamma^{(1,1)}(x_1x_2,\tau)|^2\, dA_1dA_2 . \end{aligned}$$

where $\langle i \rangle$ is the mean current in the photodetector.

To determine the effect of the finite detector area on the last term in eq. 3.4.6 we use the coherent mode description to second order by insertion of eq. 3.1.7 in this term:

$$\int_A\int_A |\Gamma^{(1,1)}(x_1x_2,\tau)|^2\, dA_1dA_2 =$$

$$A^2 \int\int \sum_{\Delta_1\Delta_2} a(\Delta_1\omega_1)a(\Delta_2,\omega_2)e^{i(\omega_1-\omega_2)\tau}\, K(\Delta_1\omega_1\Delta_2\omega_2)d\omega_1\, d\omega_2, \qquad (3.4.7)$$

where A is the detector area and

$$K(\Delta_1\omega_1\Delta_2,\omega_2) = \left| \frac{1}{A}\int U^*_{\Delta_1\omega_1}(x)\, U_{\Delta_2,\omega_2}(x)\, dx \right|^2 \qquad (3.4.8)$$

is a weight factor, which describes the interaction of the two modes $(\Delta_1\omega_1)$ and (Δ_2,ω_2) at the detector surface. The weight factor K will here be called the detector coherence factor, with the theory of optical coherence in mind, although in this case we are dealing with quantities that are deterministic. The detector coherence factor defines a coherence "volume" in the mode space seen from the detector point of view. If this mode volume includes all the excited modes in the light beam, e.g. when the detector is a point detector, the detector will weight the modes equally and the detection

process is called coherent. In the opposite case, where only one mode is present in the mode volume, the detection process is incoherent.

If the light beam is stationary in space, we can give an explicit expression for the detector coherence factor. The detector is assumed circular with a radius a, and its surface is placed in the x-y plane.

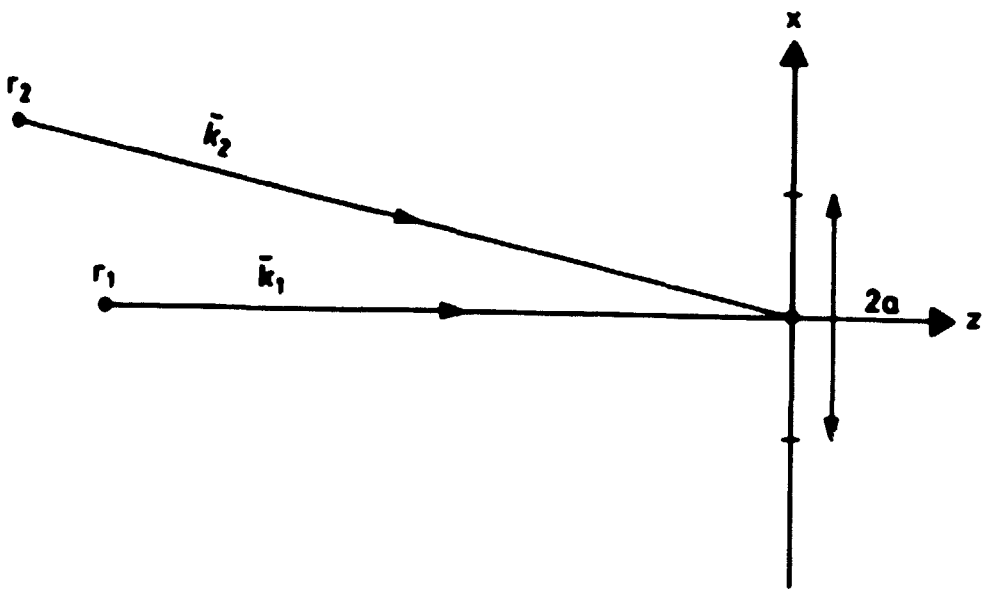


Fig. 3.3

For the detector coherence factor, we then obtain:

$$K(\omega_1 \bar{k}_1 \omega_2 \bar{k}_2) = \left| \frac{1}{A} \int e^{i(\bar{k}_2 - \bar{k}_1) \cdot \bar{x}} \, dA \right|^2 = \left\{ \frac{2J_1(\Delta ka)}{\Delta ka} \right\}^2 \qquad (3.4.9)$$

where J_1 is a Bessel function and

$$\Delta k = | \bar{z} \times (\bar{k}_1 - \bar{k}_2) | .$$

The mode volume can be defined by the relation

$$\frac{2J_1(\Delta ka)}{\Delta ka} = \frac{1}{2}$$

which gives, approximately,

$$\Delta ka \lesssim \frac{\pi}{2} . \qquad (3.4.10)$$

To interpret eq. 3.4.10, we will consider a case where $\bar{k}_1$ is a mode in the z-direction. Modes in the mode coherence volume centred around $\bar{k}_1$ are then modes where

$$(k_x^2 + k_y^2)^{1/2} < \frac{\pi}{2a}$$

Another way to interpret eq. 3.4.10 is to go back to the real space. In fig. 3.3 the two point sources are placed in the space point r_1 and r_2. If we assume that the point source is located far from the detector, i.e.

$$r_1, r_2 \gg a, \tag{3.4.11}$$

then the fields at the detector can be approximated with plane waves, and the fields from the two point sources will give two excitations in the mode space, which can be denoted $\bar{k}_1$ and $\bar{k}_2$. $\bar{k}_1$ and $\bar{k}_2$ are given by:

$$\bar{k}_1 = k_1 \frac{\bar{r}_1}{r_1} \quad \text{and} \quad \bar{k}_2 = k_2 \frac{\bar{r}_2}{r_1}.$$

From relation 3.4.10, we then obtain:

$$\left| \hat{z} \times \left(k_1 \frac{\bar{r}_1}{r_1} - k_2 \frac{\bar{r}_2}{r_2} \right) \right| \leq \frac{\pi}{2a}$$

which define a coherence volume in space. In the case where r_1 is placed at the z-axis (see fig. 3.3), we obtain from eq. 3.4.12

$$(x_2^2 + y_2^2)^{1/2} \leq \frac{\pi}{2ak_2} r_2, \tag{3.4.13}$$

and the volume of coherence is then a cone. In areas which fulfil the condition stated in eq. 3.4.11, eq. 3.4.13 is equivalent to the antenna theorem[13].

4. QUANTUM-MECHANICAL DESCRIPTION OF LIGHT

4.1. Introduction

The classical description of light can explain a wide class of experiments in optics, especially if we apply a mixed description in cases where light interacts with matter, i.e. the atoms are described quantum-mechanically and the light classically. For a certain class of phenomena, the explanation

lies outside the conceptual basis of the classical theory of light. The photoelectric effect and photon counting experiments (which will be described in chapter 5) can only be understood with a particle concept of light.

Historically, the concept of light as particles goes back to Newton's corpuscular theory of light, but in the following centuries his theory was rejected and replaced by the Huygens concept of light as waves. The reason for this rejection lay in the diffraction experiments carried out by Young and Fresnel. From a pure particle point of view, it was impossible that particles could be diffracted, and throughout the 19th century the corpuscular theory of Newton was entirely forgotten. The quantum theory of light was proposed in 1900 by Planck[14], who postulated that the energy of a harmonic oscillator, oscillating with the frequency ν, was quantized with the fundamental energy quantum $h\nu$. The constant h was named Planck's constant. Using this postulate, he could explain measurements of the spectral distribution of energy radiated from a thermal light source. Later (1905) Einstein[15] explained the photoelectric effect, which was outside a classical understanding, by assuming corpuscularity of the electromagnetic radiation. The energy of radiation was quantized in quantum of the energy $h\nu$ where ν was the frequency of the light. This fundamental energy quantum was first later (1926) named a photon. There was still an apparent incompatibility between the wave and the corpuscular point of view when Einstein explained the photoelectric effect. First with N. Bohr's interpretation of quantum mechanics was it possible to get a coinsistent picture of light explaining both diffraction phenomena and the photoelectric effect. In a purely mechanical concept of particles, the trajectory of a particle is deterministic, if we know the initial position of the particle and the force acting on the particles. In the Bohr interpretation of quantum mechanics, the trajectory of a particle is encumbered with a principal uncertainty that is related to an uncertainty in the momentum of the particle by the Heisenberg uncertainty relation:

$$\Delta r \cdot \Delta p = \hbar = h/2\pi ,$$

This principal uncertainty is the result of the quantum mechanical description, which describes the particle position as a probability wave. In the case of photons, the momentum is $\hbar\vec{k}$, where $\vec{k}$ is the wavevector of the light, and we obtain the uncertainty relation

$$\Delta r \cdot \Delta k = 1 ,$$

which expresses that the uncertainty in the wavevector of the photon will always be greater than $1/\Delta r$, i. e. the photon is able to be diffracted as a wave, so we can conclude that there is no incompatibility between the wave and particle concept in the quantum concept of particles.

The basis for a quantum description of the electromagnetic field is the Maxwell equation, which is equivalent to the equation of motion in ordinary particle mechanics. In the general case where an electric charge is present, the Maxwell equation can be deduced from a Lagrangian density function, which depends on the position of the charges and the magnetic vector potential of the field. From the Lagrangian function we can find the classical Hamiltonian function for the system[16]. By "translations" of this classical Hamiltonian, i. e. replacing the relevant quantities by operators, we obtain the quantum mechanical Hamilton operator, and further by using the so-called "second quantization" technique, we can express the Hamilton operator in terms of operators related to the photon concept. In this chapter we will only give a quantum mechanical description of the free electromagnetic field (e. g. in the absence of charges) in vacuum, and refer to textbooks for the general case[16,17].

4. 2. Quantum Description of the Fields

The Maxwell equation for the free electromagnetic field in vacuum is

$$\nabla \times \bar{H} = \frac{\delta \bar{D}}{\delta t}$$

$$\nabla \times \bar{E} = -\frac{\delta \bar{B}}{\delta t}$$

$$\nabla \cdot \bar{B} = 0$$

$$\nabla \cdot \bar{E} = 0$$

with the relations,

$$\bar{D} = \varepsilon_0 \bar{E} \quad \text{and} \quad \bar{H} = 1/\mu_0 \bar{B},$$

where ε_0 and μ_0 are the electric permittivity and magnetic permeability in vacuum.

In the absence of charges we can, by introducing the magnetic vector potential $\bar{A}$, express the electric and magnetic fields $\bar{E}$ and $\bar{B}$ as

$$\bar{E} = -\frac{\delta \bar{A}}{\delta t}$$

and

$$\bar{B} = \nabla \times \bar{A}.$$

The Maxwell equation is then reduced to

$$\nabla^2 \bar{A} - \mu_o \epsilon_o \frac{\delta^2 \bar{A}}{\delta t^2} = 0 \qquad (4.2.1)$$

where we used the London gauge[17]

$$\nabla \cdot A = 0 . \qquad (4.2.2)$$

To find a solution to eq. 4.2.1, we consider a rectangular region in space with the side lengths L_x, L_y and L_z, and apply periodical boundary conditions of the vector potential to this cavity. With this, the solution to eq. 4.2.1 can be written

$$\bar{A}(r,t) = \frac{1}{V^2} \sum_{\bar{k}} A_k(t)\, e^{i\bar{k}\cdot\bar{r}} + A_k^*(t)\, e^{-i\bar{k}\cdot\bar{r}} \qquad (4.2.3)$$

where $V = L_x L_y L_z$ is the volume of the cavity. According to the condition of periodical boundaries the wave vector $\bar{k}$ must fulfil the condition:

$$k_x = \frac{2\pi}{L_x} n_x \quad k_y = \frac{2\pi}{L_y} \quad b_z = \frac{2\pi}{L_z} n_z \qquad (4.2.4)$$

where n_x, n_y and n_z are integers.

By insertion of eq. 4.2.4 into eq. 4.2.1 we obtain

$$\ddot{A}_k + \omega_k^2 \bar{A}_k = 0 \qquad (4.2.5)$$

where $\omega_k = ck$.

Equation 4.2.5 is the differential equation of the harmonic oscillator which has the solution

$$\bar{A}_k(t) = \bar{A}_k\, e^{-i\omega_k t} .$$

The London gauge, eq. 4.2.2, gives two independent directions for $\bar{A}_k$, so the vector potential can be written

$$\bar{A}(r,t) = \frac{1}{V^{1/2}} \sum_{\bar{k}} \sum_{s=1}^{2} \bar{e}_{ks} A_{ks} e^{i(\bar{k}\cdot\bar{r} - \omega_k t)} + \bar{e}_{ks} A_{ks}^* e^{-i(\bar{k}\cdot\bar{r} - \omega_k t)} \qquad (4.2.6)$$

where $\bar{e}_{ks}$ is a unit vector which must fulfil the condition:

$$\bar{k} \cdot \bar{e}_{ks} = 0.$$

Now, considering the properties of A_{ks} and A^*_{ks} from a quantum mechanical viewpoint, it is suitable to introduce a generalized mode "position" Q_{ks} and mode "momentum" P_{ks} by the transformation

$$A_{ks} = (4\varepsilon_o\omega_k^2)^{1/2}(\omega_k Q_{ks} + i P_{ks}) . \tag{4.2.8}$$

The mode "position" Q_{ks} and the mode momentum P_{ks} satisfy the harmonic oscillator equation 4.2.5, and we can express the energy of this harmonic oscillator with a unit mass by:

$$H_{ks} = 1/2(P_{ks}^2 + \omega_k^2 Q_{ks}^2) . \tag{4.2.9}$$

The generalized "position" and "momentum" are canonically conjugated variables and, classically, we obtain the equation of motion by the relations:

$$\dot{P}_{ks} = -\frac{\delta \mathcal{H}_{ks}}{\delta Q_{ks}}$$

and

$$Q_{ks} = \frac{\delta \mathcal{H}_{ks}}{\delta P_{ks}}$$

which give the equation of the harmonic oscillator.

In quantum mechanics Q_{ks} and P_{ks} are operators, and with the usual translation we have

$$Q_{ks} \rightarrow \hat{Q}_{ks}$$

$$P_{ks} \rightarrow \hat{P}_{k_s} = \frac{\hbar}{i}\frac{\delta}{\delta Q_{ks}} .$$

The commutator of $\hat{Q}_{ks}$ and $\hat{P}_{qr}$ becomes:

$$[\hat{Q}_{ks}, \hat{P}_{qr}] = i\hbar\, \delta_{q,k}\delta_{s,r}$$

By using eq. 4.2.8 we can obtain the commutator relation for $\hat{A}_{ks}$ and $\hat{A}^*_{ks}$

$$[\hat{A}_{ks}, \hat{A}_{pr}] = 0 \tag{4.2.10}$$

and

$$[\hat{A}_{ks}, \hat{A}_{qr}] = \frac{\hbar}{2\varepsilon_o\omega_k}\,\delta_{k,q}\delta_{s,r} . \tag{4.2.11}$$

For convenience, we normalize the $\hat{A}_{ks}$'s by introducing

$$\hat{a}_{ks} = \left(\frac{\hbar}{2\epsilon_o \omega_k}\right)^{-1/2} \hat{A}_{ks} \tag{4.2.12}$$

and

$$\hat{a}^{+}_{ks} = \left(\frac{\hbar}{2\epsilon_o \omega_k}\right)^{-1/2} \hat{A}^{*}_{ks} . \tag{4.2.13}$$

From eqs. 4.2.10 and 4.2.11 we obtain

$$[\, \hat{a}_{ks}, \hat{a}_{qr} \,] = 0 \tag{4.2.14}$$

and

$$[\, \hat{a}_{ks}, \hat{a}^{+}_{qr} \,] = \delta_{k,q} \delta_{s,r} . \tag{4.2.15}$$

The energy of the k^{th} mode of the harmonic oscillator can then be expressed:

$$\hat{\mathcal{H}}_{ks} = \hbar\omega_k (\hat{a}^{+}_{ks} \hat{a}_{ks} + 1/2) . \tag{4.2.16}$$

Now, as is known, the eigenvalues of the Hamiltonian for the harmonic oscillator constitute a discrete spectrum given by the relation

$$E_{n_{ks}} = \hbar\omega_k (n_{ks} + 1/2) \tag{4.2.17}$$

where n_{ks} is a positive integer.

The eigenstates of $\hat{\mathcal{H}}_{ks}$ constitute an orthonormal and complete set, and we denote the eigenstates as number states or the Fock state. The number states are defined by the relation

$$\hat{\mathcal{H}}_{ks} \, | n_{ks} \rangle = E_{ks} \, | n_{ks} \rangle . \tag{4.2.18}$$

The number n_{ks} is defined to indicate a number of "particles" - photons - or a number of excitations in the mode ks. With this concept the operator $\hat{a}^{+}_{ks} \hat{a}_{ks}$ can be interpreted as a number operator, i. e. an operator for the number of photons with the wave vector k

$$\hat{n}_{ks} = \hat{a}^{+}_{ks} \hat{a}_{ks} . \tag{4.2.19}$$

From eqs. 4.2.14, 4.2.15 and 4.2.16 we find the effect of the number operator on the number state

$$\hat{n}_{ks} | n_{ks} \rangle = n_{ks} | n_{ks} \rangle ,$$

i.e. the number states are eigenstate for the number operator. To find the effect of the operators $\hat{a}^{+}_{ks}$ and $\hat{a}_{ks}$, we use the commutator relations 4.2.12 and 4.2.13 to obtain

$$\hat{n}_{ks}(\hat{a}^{+}_{ks} | n_{ks} \rangle) = [\hat{n}_{ks}, \hat{a}^{+}_{ks}] | n_{ks} \rangle + n_{ks}(\hat{a}^{+}_{ks} | n_{ks} \rangle) = (n_{ks}+1)(\hat{a}^{+}_{k} | n_{ks} \rangle),$$

i.e. the state $\hat{a}^{+}_{k} | n_{ks} \rangle$ is an eigenstate of n_{ks} with the eigenvalue $n_{ks}+1$, which means that

$$\hat{a}^{+}_{ks} | n_{ks} \rangle \propto | n_{ks}+1 \rangle . \qquad (4.2.20)$$

In the same way we obtain for a_{ks}

$$\hat{a}_{ks} | n_{ks} \rangle \propto | n_{ks}-1 \rangle \qquad n_{ks} \geq 1$$

$$\hat{a}_{ks} | n_{ks} \rangle = 0 \qquad n_{ks} = 0$$

The effect of $\hat{a}^{+}_{ks}$ and $\hat{a}_{ks}$ will then be defined by the relations according to eqs. 4.2.18 and 4.2.19,

$$\hat{a}^{+}_{ks} | n_{ks} \rangle = (n_{ks}+1)^{1/2} | n_{ks}+1 \rangle \qquad (4.2.22)$$

and

$$\hat{a}_{ks} | n_{ks} \rangle = n_{ks}^{1/2} | n_{ks}-1 \rangle , \qquad (4.2.23)$$

i.e. $\hat{a}^{+}$ and $\hat{a}$ can be interpreted as a creating operator and a destructing operator, respectively. $\hat{a}^{+}_{ks}$ creates an excitation or a photon in the mode ks, and $\hat{a}_{ks}$ destroys an excitation or a photon in the mode ks. Corresponding to the mode operators $\hat{a}^{+}$ and $\hat{a}$, we can define the operators

$$\hat{\Psi}^{+}(r,t) = \frac{1}{V^{1/2}} \sum_{k,s} \bar{e}_{ks} \hat{a}^{+}_{ks} e^{-i\bar{k}\cdot\bar{r}+i\omega_k t} \qquad (4.2.24)$$

and

$$\hat{\Psi}^{-}(r,t) = \frac{1}{V^{1/2}} \sum_{k,s} \bar{e}_{ks} \hat{a}_{ks} e^{i\bar{k}\cdot\bar{r}-i\omega_k t} \qquad (4.2.25)$$

and as $\hat{a}^+_{ks}$ creates a photon in the mode k_s, $\hat{\Psi}^+(r,t)$ can be interpreted as an operator that creates a photon in the space-time point $(\bar{r},t)$. Furthermore, the product

$$\hat{\Psi}^+(r,t)\hat{\Psi}^-(r,t) \tag{4.2.26}$$

gives the number density of the photons at the space-time point $(\bar{r},t)$. From eqs. 4.2.24 and 4.2.25 we see that $\hat{\Psi}^+$ and $\hat{\Psi}^-$ are closely related to the field quantities $\bar{E}$, $\bar{B}$ and $\bar{A}$.

The reason for introducing the quantities $\hat{\Psi}^+$ and $\hat{\Psi}^-$ here is to point out the similarity between the number density expressed in eq. 4.26 and the usual probability density $|\psi(r,t)|^2$ in ordinary quantum mechanics for a single particle. Further, application of the formal technique called "second quantization" to a system of many indistinguishable particles yields a description similar to the description of the light field by photons. The "particles" in the "second quantization" will be a collective excitation of the systems (phonon, polarons, magnons, etc.), and the "particle field" can be described by quantities like $\hat{\Psi}^+$ and $\hat{\Psi}^-$ as in the case of the electromagnetic field.

From eqs. 4.2.6, 4.2.12 and 4.2.13 we can then write the vector potential in terms of the creating and destructing operators $\hat{a}^+$, $\hat{a}$:

$$\hat{\bar{A}}(\bar{r},t) = \sum_{k,s} \left(\frac{\hbar}{2\varepsilon_0 V\omega_k}\right)^{1/2} \bar{e}_{ks}\left(\hat{a}^+_{ks} e^{-i\bar{k}\cdot\bar{r}+i\omega_k t} + \hat{a}_{ks} e^{+i\bar{k}\cdot\bar{r}-i\omega_k t}\right) \tag{4.2.27}$$

and further the $\bar{E}$ field

$$\hat{\bar{E}}(\bar{r},t) = i \sum_{k,s} \left(\frac{\hbar\omega_k}{2\varepsilon_0 V}\right)^{1/2} \bar{e}_{ks}\left(\hat{a}^+_{ks} e^{-i\bar{k}\cdot\bar{r}+i\omega_k t} - \hat{a}_{ks} e^{i\bar{k}\cdot\bar{r}-i\omega_k t}\right). \tag{4.2.28}$$

Finally we here calculate the electromagnetic energy in the cavity when the field is excited to the number state

$$|\{n_{ks}\}\rangle = |n_{ks_1}\rangle\,|n_{ks_2}\rangle \ldots\ldots$$

The electromagnetic energy in a volume V is

$$H = 1/2\int_V(\bar{E}\cdot\bar{D}+\bar{H}\cdot\bar{B})dV = 1/2\,\varepsilon_0\int\left(\frac{\partial\bar{A}}{\partial t}\right)^2 + c^2(\nabla\times\bar{A})^2)dV\,, \tag{4.2.29}$$

and by using eq. 4.27 we find that

$$\langle \{n_{ks}\} | \hat{H} | \{n_{ks}\} \rangle = \sum_{k,s} \hbar\omega_k (n_{ks} + 1/2) . \qquad (4.2.30)$$

From eq. 4.2.30 we see that the energy of the electromagnetic field contains a divergent term, the zero point energy, given by

$$H_0 = 1/2 \sum_{k,s} \hbar\omega_k = \sum_{k} \hbar\omega_k ,$$

which is present even when there are no excited photons.

Now, infinite energies are conceptually difficult to deal with, and this could seem to be an "unphysical" property of the description. However, this problem is known elsewhere in physics, for instance the self-energy of a point charge; moreover, practical measurements will always give results that involve changes in the electromagnetic energy.

4.3. Coherent States of the Electromagnetic Field

The definition of coherent fields in quantum mechanics is the same as given in chapter 2 for the classical field, i.e. the correlation function of the field must factorize, as expressed by eq. 2.3.1. In order to have correlation functions that are measurable quantities, we must define our quantum mechanical correlation functions in proper accordance with the way the detection process works. In this report we only deal with the absorbing processes of photons and the way in which the photodetector operates. With respect to the more general concepts of quantum counters, we refer to a textbook in the field[18].

From eq. 4.2.28 we see that the E field can be split into two parts, one $\hat{\vec{E}}^-$, which annihilates photons, and one $\hat{\vec{E}}^+$, which creates photons:

$$\hat{\vec{E}} = \hat{\vec{E}}^+ + \hat{\vec{E}}^-$$

where

$$\hat{\vec{E}}^+ = i \sum_{k,s} \left(\frac{\omega_k}{2\varepsilon_0 V}\right)^{1/2} \vec{e}_{ks} \hat{a}^+_{ks} e^{-i\vec{k}\cdot\vec{r} + i\omega_k t} \qquad (4.3.1)$$

and

$$\hat{\vec{E}}^- = -i \sum_{ks} \left(\frac{\omega_k}{2\varepsilon_0 V}\right)^{1/2} \vec{e}_{ks} \hat{a}_{ks} e^{i\vec{k}\cdot\vec{r} - i\omega_k t} \qquad (4.3.2)$$

It is seen that $\hat{E}^+$ and $\hat{E}^-$ are equivalent to the positive and the negative frequency part of the E field introduced in chapter 2. When a photodetector records a photon, it is equivalent to an annihilation of a photon from an electromagnetic field, and the field will make a transition from an initial state $|i\rangle$ to a final state $|f\rangle$. If we let the photon be polarized in the z-direction, then the matrix element involved in the transition takes the form

$$\langle f| \hat{E}_z^-(r,t) |i\rangle \ .$$

In order not to go into the details of photo-absorption, we will assume an ideal detector, i.e. one having a frequency-independent transition rate. We then find that the probability per unit time to record a photon, in accordance with Fermi's golden Rule, is proportional with:

$$\sum_f |\langle f| \hat{E}_z^-(r,t) |i\rangle|^2 \qquad (4.3.3)$$

where the summation is extended over all possible final states. Now, the set of final states will be a complete set and we can write eq. 4.3.3

$$\sum_f |\langle f| \hat{E}_z^-(r,t) |i\rangle|^2 = \langle i| \hat{E}_z^+ \sum_f |f\rangle\langle f| \hat{E}_z^-|i\rangle = \langle i| \hat{E}_z^+(r,t)\hat{E}_z^-(r,t) |i\rangle \qquad (4.3.4)$$

where we use the completeness of the final states by using the relation

$$\sum_f |f\rangle\langle f| = 1 \ . \qquad (4.3.5)$$

The current in the photodetector will be proportional to the probability transition rate (eq. 4.3.4), so this will be a measurable quantity. In a general arrangement of n photodetectors, we find that the n-fold probability transition rate is proportional to

$$\langle i| \hat{E}_z^+(r_1,t) \ldots \hat{E}_z^+(r_n,t_n)\hat{E}_z^-(r_1,t_1) \ldots \hat{E}_z^-(r_n,t_n) |i\rangle \ . \qquad (4.3.6)$$

Now from eqs. 4.3.4 and 4.3.5 we see that measurable quantitites in quantum mechanics imply a certain order of the field operators with respect to the detection process with which we deal. When the detection is an absorption of photons, the arrangement of the operators is said to be in normal order, i.e. the quantum correlation function $\Gamma_N^{(n,m)}$ in normal order becomes:

$$\Gamma_N^{(n,m)}(\bar{r}_1,t_1\ldots\bar{r}_n t_n,\ \bar{r}_{n+1}t_{n+1}\ldots\bar{r}_{n+m},t_{n+m}) = \tag{4.3.7}$$

$$\langle i|\hat{E}_z^+(\bar{r}_1,t_1)\ldots\hat{E}_z^+(r_n,t_n)\hat{E}_z^-(\bar{r}_{n+1},t_{n+1})\ldots\hat{E}_z^-(\bar{r}_{n+m},t_{n+m})|i\rangle$$

The condition of full coherence implies that the correlation function $\Gamma_N^{(n,m)}$ be factorized, and a way to fulfil this condition is to find states of the electromagnetic field which are eigenstates of the destructing operator E^-, i.e.

$$E^-(\bar{r},t)|i\rangle = \epsilon(\bar{r},t)|i\rangle\ . \tag{4.3.8}$$

If such a state exists and the field is excited to such a state $\Gamma_N^{(n,m)}$ will factorize

$$\Gamma_N^{(n,m)} = \epsilon^*(n,t_1)\ldots\ \epsilon^*(r_n,t_n)\,\epsilon(r_{n+1},t_{n+1})\ldots\epsilon(r_{n+m},t_{n+m}).$$

The operators E^- and E^+ are non-Hermitian operators, so we cannot be sure that such eigenstates exist. By insertion of eq. 4.3.2 into the eigenvalue equation 4.3.7, we see that eq. 4.3.7 implies that the eigenstates must be eigenstates of the annihilating operator $\hat{a}_{ks}$. Denoting the eigenstates of $\hat{a}_{ks}$ by $|a_{ks}\rangle$, we find

$$\hat{a}_{ks}|a_{ks}\rangle = a_{ks}|a_{ks}\rangle\ . \tag{4.3.9}$$

The solution of eq. 4.3.8 can be found in terms of the number state $|n_{ks}\rangle$ and we obtain

$$|a_{ks}\rangle = e^{-\frac{1}{2}|a_{ks}|^2}\sum_{n_{ks}}\frac{a_{ks}^n}{(n_{ks}!)^{\frac{1}{2}}}|n_{ks}\rangle \tag{4.3.10}$$

where a_{ks} can be any complex number. The states are normalized,

$$\langle a_{ks}|a_{ks}\rangle = e^{-|a_{ks}|^2}\sum_{n_s}\frac{a_{ks}^n\,a_{ks}^{*n}}{n_{ks}!} = 1$$

but not orthogonally due to the non-Hermetian nature of a_{ks}. The matrix element $\langle a_{ks}|\beta_{ks}\rangle$ gives

$$\langle \alpha_{ks} | \beta_{ks} \rangle = e^{-\frac{1}{2}(|\alpha_{ks}|^2 + |\beta_{ks}|^2 + 2\alpha^*_{ks}\beta_{ks})}$$

The physical meaning of the complex parameter α_{ks} can be found by calculation of the number of photons in the state $|\alpha_{ks}\rangle$

$$\langle \alpha_{ks} | \hat{n}_{ks} | \alpha_{ks} \rangle = |\alpha_{ks}|^2$$

i.e. the absolute square of α_{ks} is the mean number of photons in the state $|\alpha_{ks}\rangle$.

The state $|\alpha_{ks}\rangle$ is called the coherent state of the field, for the reason that the state

$$|\{\alpha_{ks}\}\rangle = |\alpha_{ks_1}\rangle|\alpha_{ks_2}\rangle \cdots\cdots$$

will be an eigenstate of E^-, and thereby assure the factorization properties of the normal order correlation function.

4.4. Statistical Description in Quantum Mechanics

The concept equivalent to the probability density in a classical connection is the density matrix operator in quantum mechanics. The expectation value of an operator $\hat{0}$ in a physical system excited to a state $|S\rangle$ is given by

$$\langle \hat{0} \rangle = \langle S | \hat{0} | S \rangle$$

which with the closure theorem (eq. 4.3.5) can be written as:

$$\langle \hat{0} \rangle = \sum_T \langle T | (|S\rangle\langle S|) \hat{0} | T \rangle \qquad (4.4.1)$$

where $|T\rangle$ is an arbitrary state.

The density matrix operator for this system is defined by

$$\hat{\rho} = |S\rangle\langle S|$$

and eq. 4.4.1 can then be put in the form

$$\langle \hat{0} \rangle = \mathrm{Tr}(\hat{\rho}\hat{0}) \qquad (4.4.2)$$

where Tr means the trace of the operator in the parenthesis.

In a more general case, where our knowledge of the excited state of the system is of a statistical nature, the state of the system must be considered as a statistical mixture of some known states, i.e. a possible state 'u' can be expressed as

$$|u\rangle = \sum_S C_S |S\rangle \tag{4.4.3}$$

where C_S is a stochastic expansion parameter.

The mean value of an operator $\hat{0}$ is then

$$\begin{aligned}\{\langle u|\hat{0}|u\rangle\}_{av} &= \sum_{S_1,S_2} \{C^*_{S_1} C_{S_2}\}_{av} \langle S_1|\hat{0}|S_2\rangle \\ &= \sum_T \langle T| \sum_{S_1,S_2} \{C^*_{S_1} C_{S_2}\}_{av} |S_2\rangle\langle S_1|\hat{0}|T\rangle \\ &= Tr(\hat{\rho}\hat{0})\end{aligned}$$

where the density matrix in this general case is expressed by

$$\hat{\rho} = \sum_{S_1,S_2} \{C^*_{S_1} C_{S_2}\}_{av} |S_2\rangle\langle S_1| \ . \tag{4.4.4}$$

The wave function S is here an arbitrary set of functions, but in general it is possible to find a set of "pure" wave functions in which the averaged quantity $\{C^*_{S_1} C_{S_2}\}_{av}$ is of diagonal form, i.e.

$$\{C^*_{S_1} C_{S_2}\}_{av} = P_S \delta_{S_1 S_2} ,$$

where P_S can be interpreted as the probability that the pure state S is present, which is seen from the fact that P_S is a positive quantity, and from the relation

$$Tr\hat{\rho} = 1 , \tag{4.4.5}$$

which can be shown by using eqs. 4.4.4 and 4.4.3.

In the pure state representation the density matrix can then be expressed as:

$$\hat{\rho} = \sum_S P_S |S\rangle\langle S| \ . \tag{4.4.6}$$

To discuss a more specific example, we will look at thermal radiation in thermal equilibrium at the temperature T. In this case the density matrix operator will have a simple relation to the electromagnetic energy, which can be found by thermodynamical consideration[19]

$$\hat{\rho} = \frac{e^{-\beta H}}{Tr\, e^{-\beta H}} \tag{4.4.7}$$

where H is the Hamiltonian of the electromagnetic field (see eq. 4.2.9) and $\beta = 1/k_b T$ is the usual Boltzmann factor. From eq. 4.2.9 we obtain

$$\hat{H} = \sum_{ks} \hbar\omega_k (\hat{n}_{ks} + \tfrac{1}{2}). \tag{4.4.8}$$

From eq. 4.4.7 we obtain the matrix element

$$\langle n_{ks} | \rho | m_{ks} \rangle = e^{-\beta\hbar\omega_{ks} n_{ks}} (1 - e^{-\beta\hbar\omega_{ks}}) \delta_{n_{ks}, m_{ks}}, \tag{4.4.9}$$

which means that the number states in this case are pure states. The probability that the state $| n_{ks} \rangle$ is present can then according to eq. 4.4.6 be expressed as

$$P(n_{ks}) = e^{-\beta\hbar\omega_{ks} n_{ks}} (1 - e^{-\beta\hbar\omega_{ks}}). \tag{4.4.10}$$

The mean number of photons with the wavevector $\bar{k}$ becomes

$$\begin{aligned} \langle \hat{n}_{ks} \rangle &= Tr(\hat{\rho}\hat{n}_{ks}) \\ &= \sum_{n_{ks}} P(n_{ks}) n_{ks} \\ &= \frac{e^{-\beta\hbar\omega_k}}{(1 - e^{-\beta\hbar\omega_k})} \end{aligned} \tag{4.4.11}$$

By insertion of eq. 4.5.11 into 4.4.10 we obtain

$$P(n_{ks}) = \frac{\langle n_{ks} \rangle^{n_{ks}}}{(1 + \langle n_{ks} \rangle)^{n_{ks}+1}}, \tag{4.4.12}$$

which is the Bose-Einstein distribution.

4.5. Glauber-Sudarshan Representation of the Density Matrix

The density matrix of the electromagnetic field will in general not have the number states as pure states, but it can be shown that the coherent states possess these properties. The proof for this is rather lengthy and will not be given here, but reference is made to papers by Glauber[20] and Sudarshan[21]. The representation of the density matrix in a diagonal form in terms of the coherent state is a remarkable property of the coherent states. The diagonal form of the density matrix gives rise to a formal equivalence between the classical and the quantum description of light, which will be shown later on. For a single mode, the Glauber-Sudarshan representation of the density matrix has the form

$$\hat{\rho} = \int P_N(\alpha) |\alpha\rangle\langle\alpha| d^2\alpha, \tag{4.5.1}$$

where the integration must be sustained over the complex plane, i.e. $d^2\alpha = d\,\mathrm{Re}\alpha\; d\,\mathrm{Im}\alpha$.

Since $\mathrm{Tr}\hat{\rho} = 1$ we obtain

$$\int P_N(\alpha) d^2\alpha = 1 \tag{4.5.2}$$

and from the Hermitidity of $\hat{\rho}$ it follows that $P_N(\alpha)$ is a real function. Meanwhile $P_N(\alpha)$ is not in general a positive function, so an interpretation of $P_N(\alpha)$ as a probability function is not possible, and we will denote it a quasi-probability function. The cases where $P_N(\alpha)$ is negative must correspond to the experiments having no classical analogue, and, contrarily, if $P_N(\alpha)$ is a positive function in the whole complex plane, then a classical interpretation of such an experiment must be possible.

4.6. Thermal Light

As an example of the quasi-probability function $P_N(\alpha)$ we find P_N in the case of chaotic light, where the density matrix has the number states as pure states, and the probability density of the number states is based on the Bose-Einstein distribution (se q. 4.4.12), i.e.

$$\hat{\rho} = \sum \frac{\langle n\rangle^n}{(1+\langle n\rangle)^{n+1}} |n\rangle\langle n|. \tag{4.6.1}$$

By using the closure theorem of the coherent state, i.e.

$$\frac{1}{\pi} \int |\alpha\rangle\langle\alpha| d^2\alpha = \hat{1}, \tag{4.6.2}$$

we obtain, using the definition of the coherent states (eq. 4.3.10),

$$\hat{\rho} = \frac{1}{\pi^2} \iint |\beta\rangle \Big(\sum_n \frac{\langle n\rangle^n}{(1+\langle n\rangle)^{n+1}} \langle \beta|n\rangle\langle n|\alpha\rangle \Big) \langle \alpha| d^2\alpha d^2\beta$$

$$= \frac{1}{\pi} \int \sum_{n,m} e^{-\beta^2} \frac{\langle n\rangle^n}{(1+\langle n\rangle)^{n+1}} \frac{(\beta^*)^n}{(n!)^{\frac{1}{2}}} \frac{(\beta)^m}{(m!)^{\frac{1}{2}}} |m\rangle\langle n| d^2\beta \ .$$

Now the terms in the sum will only give contribution for $n = m$, so by using the substitution

$$\beta = (1+\frac{1}{\langle n\rangle})^{\frac{1}{2}}\alpha \ ,$$

we obtain

$$\hat{\rho} = \int |\alpha\rangle\langle\alpha| \frac{1}{\pi\langle n\rangle} e^{-|\alpha|^2/\langle n\rangle} d^2\alpha$$

and with it

$$P_N(\alpha) = \frac{1}{\pi\langle n\rangle} e^{-|\alpha|^2/\langle n\rangle} \ . \qquad (4.6.3)$$

4.7. The Ideal Laser

As a second example of the quasi-probability function $P_N(\alpha)$ we look at the field excitation from an ideal laser. In a laser medium the atoms or molecules are pumped from an energetic ground level to an excited state by some outer pump mechanism. The atoms in the excited level will undergo transitions to an intervening level by the emission of photons with an energy corresponding to the energy difference between the two levels of the atomic system. The emission of light can take place in two ways, either spontaneously (i. e. uncorrelated with the emission from the remaining atoms and the light field present) or as stimulated emission, an effect first predicted by Einstein (1905)[22]. The probability for a stimulated emission of a photon is proportional to the number of photons of the same kind that are present in the medium. The stimulated emission gives rise to the laser effect; the more a single mode is present, the more photons are emitted in the same mode, which means that the excited mode will be coherent. Now, which of the modes is excited depends on the geometry of the walls surrounding the medium and the possible transitions in the atomic

system. The growth of a possible mode is limited by different loss mechanisms in the medium, by the pump rate and further by the growth of other possible modes.

In a model of an ideal laser we assume that only one mode is present. The present mode is not strictly coherent due to the effect of the spontaneous emission, and this effect can be considered as a noise contribution. The quasi-probability function for the coherent state $|\alpha\rangle$ is given by

$$P_n^{(1)}(\beta) = \delta(\alpha-\beta) \tag{4.7.1}$$

which, by introducing the amplitude and phase of α and β, can be written as

$$P_n^{(1)}(\beta) = \delta(|\alpha|-|\beta|)\, e^{i(\phi_\beta-\phi_\alpha)}, \tag{4.7.2}$$

The effect of the spontaneous emission can be considered as a lack of knowledge of the phase φ_α, so the actual quasi-probability distribution for an ideal laser is an uniform averaging of eq. 4.7.2 in the interval $(0\ 2\pi)$ of the phase φ_α, i.e.

$$\begin{aligned} P_N(\beta) &= \frac{1}{2\pi}\int_0^{2\pi} \delta(|\alpha|-|\beta|)e^{i(\phi_\beta-\phi_\alpha)}d\varphi_\alpha \\ &= \frac{1}{\pi}\delta(|\alpha|^2-|\beta|^2) \end{aligned} \tag{4.7.3}$$

and the density matrix then becomes

$$\hat{\rho} = \frac{1}{\pi}\int\delta(|\alpha|^2-|\beta|^2)\,|\beta\rangle\langle\beta|\,d^2\beta. \tag{4.7.4}$$

The matrix element of ρ in terms of the number state becomes

$$\langle n|\hat{\rho}|m\rangle = \delta_{n,m}\,\frac{(|\alpha|^2)^n}{n!}\,e^{-|\alpha|^2} \tag{4.7.5}$$

which shows that the number of photons in a single laser mode has a Poisson distribution.

The laser model stated here is rather primitive, and to obtain a more realistic picture of laser action we refer to models presented by Glauber[23] and H. Haken[24].

4.8. Relation Between the Quantum and Classical Descriptions of Light

The relations between the quantum and classical descriptions of light can shortly be stated as an equivalence between the normal order quantum correlation functions and the corresponding classical correlation functions, by finding the classical probability density in terms of the quasi-probability function. The classical probability density becomes:

$$P_n(V_1^+ \ldots V_n^+, V_1^- \ldots . V_n^-) =$$

$$\mathrm{Tr}(\hat{\rho} :\Pi(\delta(V_i^+ - \hat{V}_i^+)\Pi\delta(\hat{V}_i^- - V_i)):) = \tag{4.8.1}$$

$$\int P_N(\{\alpha_k\})\ \Pi\delta(U_i^+ - V_i^+)\delta(U_i^- - V_i^-)d^2\{\alpha_k\}$$

where

$$V_i^+ = V^+(\bar{r}_i, t_i)$$

and U_i^+ is given by the relation (see eq. 4.3.8)

$$\hat{V}_i^+|\{\alpha_k\}\rangle = U_i^+|\{\alpha_k\}\rangle .$$

No examples of this transformation are given here, but if, for instance, we wish to transform the quasi-probability density for chaotic light (eq. 4.6.3), a simple way to make the calculation is to use the characteristic function of $P_n(V)$. As can be expected, the result gives a Gaussian distribution of the fields.

5. PHOTON COUNTING

5.1. Photon Counting Equipment and the Photo Detecting Process

The quantum concept of light as photons gives rise to a new type of experiment, photon counting, that is new in respect to the conventional interference and spectroscopic methods of investigating the properties of light. Photon counting gives information on the statistics of a given light source, and especially measures the coherence time. The technique of measuring coherence time is called intensity-fluctuation spectroscopy and it constitutes a useful tool for measuring spectral widths from 1 to 100 MHz. Using conventional methods, one can measure spectral widths down to 10 MHz (e.g. with a Fabry-Perot interferometer).

An experimental set-up to measure the statistical properties of a light source is sketched in fig. 5.1.

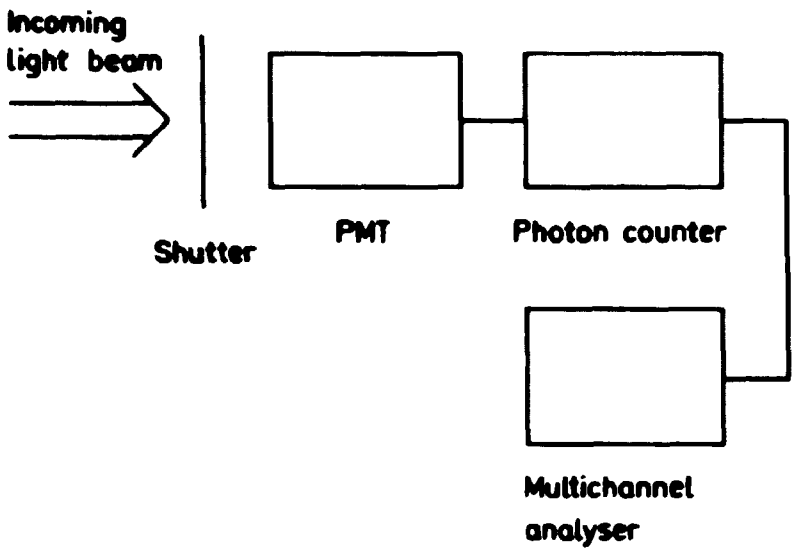


Fig. 5.1

The incoming light is detected by a photomultiplier (PMT) connected to a photon counter. The photon counter records the number of photo electrons from the PMT, in a time interval of length T determined by the shutter in front of the PMT. The measured number of counts is then recorded by the multichannel analyzer. After a certain time, longer than the coherence time of the light, the shutter is reopened and the measurement is repeated. The reason for waiting for a time longer than the coherence time of the light, is that we wish to obtain independent samples. After a sufficient number of measurements, we can find a distribution for the number of counts in the time interval T.

A survey of the way in which the PMT works gives a more detailed picture of the detection process. A photon hitting the cathode of the PMT will give rise to the emission of an electron. The probability, $p(t)\Delta t$, that an electron is emitted in a time interval Δt will be proportional to the number of photons per time unit hitting the cathode and with the time interval Δt, i. e.

$$p(t)\Delta t = \xi\, n(t)\Delta t\,, \qquad (5.1.1)$$

where the constant ξ is the quantum efficiency of the detector, its value depending on the cathode material and the frequency of the incoming light.

The emitted electron is then multiplied, so that a current pulse can be measured as an output of the PMT. The multiplicator consists of a chain of dynodes. If a dynode is hit by an electron it causes the emission of a cascade of electrons. The emitted electrons are then accelerated by an electric field so they hit the next dynode. If the mean number of electrons emitted from a dynode, hit by a single electron, is μ, then the mean number of electrons emitted from the n^{th} dynode will be μ^n. In commercially available PMT's an amplification as high as 10^8 is obtainable.

The multiplication process can be wholly characterized by a pulse-height distribution. The current impulse from the PMT resulting from a single incoming photon is essentially a stochastic quantity. Another effect that disturbs the pulse from the PMT is the dark current, which results from the thermal electrons emitted from the dynodes and the cosmic background radiation that causes the emission of electrons from the cathode. The thermal dark current pulse-height dominates at small pulse heights, whereas the contribution from cosmic radiation lies at larger pulse heights. The thermal dark current is strongly temperature dependent and can be reduced by cooling the PMT.

To overcome the effect of the dark current in the PMT, an upper and lower discrimination level can be introduced at the output of the PMT to assure that a detected current pulse is really caused by a photon. In the following it is assumed that the effect of the discriminator levels and the stochastic nature of the current pulses is incorporated in the quantum efficiency ξ (see eq. 5.1.1).

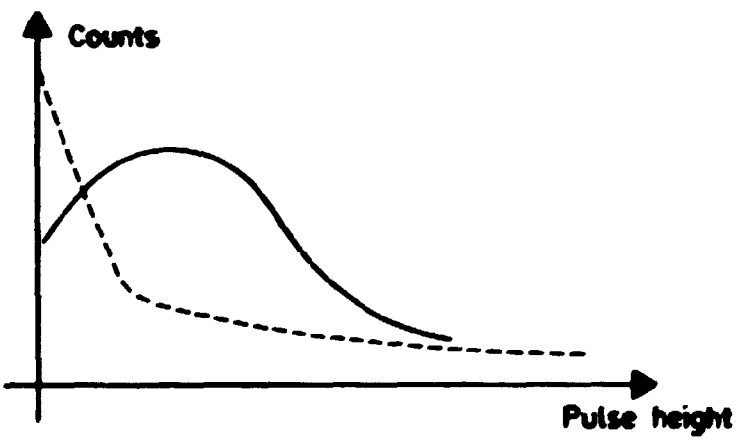


Fig. 5.2

5.2. Photon Counting Statistics

The photon is a quantum-mechanical concept, and to find the distribution function of the number of photo electrons counted in a time interval T, it would be proper to use a full quantummechanical treatment as done by Kelly and Kleiner (1968)[25]. The quantum treatment is rather lengthy and is not used here. Instead, we use the semi-classical approach of Mandel (1958)[26]. where the light is treated classically and the detection process is treated quantum-mechanically.

By assuming that different photo-electric events are statistically independent, and by using a first-order perturbation theory with respect to the effect of interaction between the electromagnetic field and the atoms in the photo detector, one can show that the probability for the photo emission of an electron in a time interval $(t/t+\Delta t)$ is

$$p(t)\Delta t = \alpha I(t)\,\Delta t, \qquad (5.2.1)$$

where I(t) is the intensity of the incoming light at the detector surface.

The photon counting experiment is started at a time t, and we find the probability for the event that m photo electrons will be counted during a time interval $(t \mid t+t' + \Delta t)$

The probability will be denoted $P_m(t \mid t'+\Delta t)$. t denotes a "small" time increment. The event that m photo-electrons have been counted in the time interval $(t \mid t+t'+\Delta t)$ can occur by counting m photo-electrons in the time interval $(t \mid t+t')$, or by counting m-1 photo electrons in the time interval $(t \mid t+t')$ and one photo-electron in the time interval $(t+t' \mid t+t'+\Delta t)$. When Δt is considered "small", we can neglect the event that two counts are recorded in the time interval $(t+t' \mid t+t'+\Delta t)$. P_m can then be expressed as:

$$P_m(t \mid t'+\Delta t) = P_m(t \mid t')(1-p(t'+t)\Delta t) + P_{m-1}(t \mid t')p(t+t')\Delta t\,. \qquad (5.5.2)$$

By letting $\Delta t \to 0$ we obtain

$$\frac{d\,P_m(t,t')}{dt'} = -p(t+t')\,(P_m(t \mid t') - P_{m-1})t \mid t')), \qquad (5.2.3)$$

which has the solution:

$$P_m(t,T) = \frac{(\alpha w)^m}{m!}\,e^{-\alpha w}, \qquad (5.2.4)$$

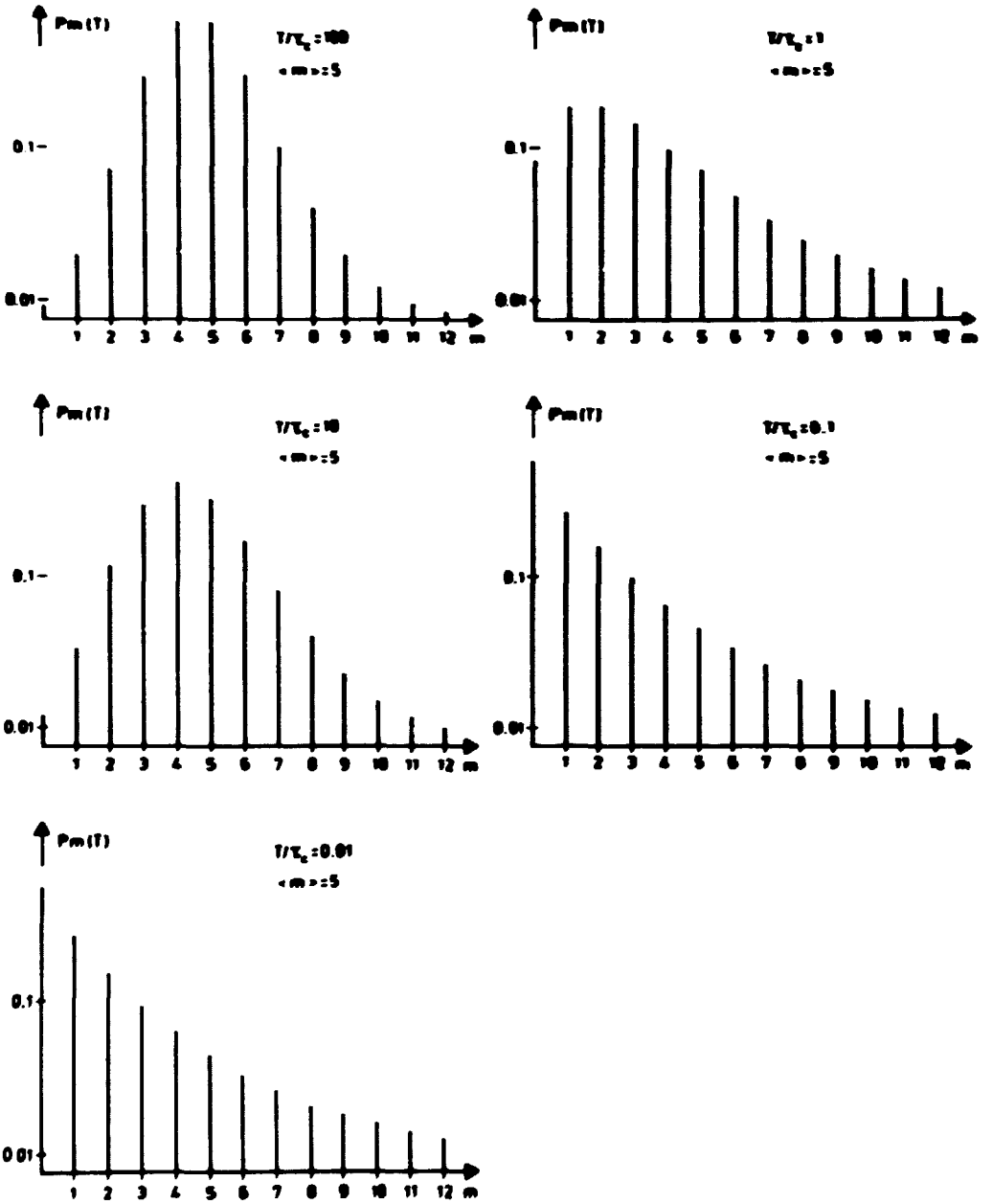

Pm(T)
T/τc :100
<m>:5
0.1
0.01
1 2 3 4 5 6 7 8 9 10 11 12 m
Pm(T)
T/τc :1
<m>:5
0.1
0.01
1 2 3 4 5 6 7 8 9 10 11 12 m
Pm(T)
T/τc :10
<m>:5
0.1
0.01
1 2 3 4 5 6 7 8 9 10 11 12 m
Pm(T)
T/τc :0.1
<m>:5
0.1
0.01
1 2 3 4 5 6 7 8 9 10 11 12 m
Pm(T)
T/τc :0.01
<m>:5
0.1
0.01
1 2 3 4 5 6 7 8 9 10 11 12 m

Fig. 5.3

where

$$w = 1/a\int_{t}^{t+T} p(t')dt' = \int_{t}^{t+T} I(t')dt'. \qquad (5.2.5)$$

The distribution function $P_m(t, T)$ represents a distribution where counting started at the time t; as the integrated intensity w is a stochastic variable we must perform an ensemble averaging to achieve a mean distribution function, i.e.

$$\langle P_m(t,T)\rangle = \int_0^\infty P(w)\frac{(aw)^m}{m!}e^{-aw}dw, \qquad (5.2.6)$$

where P(w) is the distribution function for the integrated intensity.

From eq. 5.2.5 we see that $P_m(t, T)$ is the Poisson-transformed of the distribution function P(w). In the case of a stationary light beam, $P_m(t, T)$ will be independent of the starting time t. In the following we will assume stationarity and further omit the averaging brackets for the sake of brevity. We can then write eq. 5.2.6 as:

$$P_m(T) = \int_0^\infty P(w)\frac{(aw)^m}{m!}e^{-aw}\,dw\ . \qquad (5.2.7)$$

The moments of $P_m(T)$ can be expressed in terms of moments of the distribution function for the integrated intensity by the equation

$$\langle m^p\rangle = \langle (e^{-x}(x\frac{d}{dx})^p e^x)\,|_{x=aw}\rangle \qquad (5.2.8)$$

which, for the first and second moment, gives

$$\langle m\rangle = \langle aw\rangle \qquad (5.2.9)$$

and

$$\langle m^2\rangle = \langle aw\rangle + \langle (aw)^2\rangle \qquad (5.2.10)$$

From eqs. 5.2.10 and 5.2.4 we find that the variance of the counts is given by

$$\langle (\Delta m)^2\rangle = \langle m\rangle + \langle (aw-\langle aw\rangle)^2\rangle \qquad (5.2.11)$$

and we see that even in the absence of fluctuations in the integrated intensity, the number of counts fluctuates. This effect is due to the discrete nature of the photo-electric effect, and consequently this term has nothing to do with the statistical properties of the light beam. The last term in eq. 5.2.11 expressed the contribution from the fluctuation in the light beam. The technique where only the second-order fluctuation is investigated is called "intensity fluctuation spectroscopy".

The equivalent quantum-mechanical expression for the probability to count m photo-electrons in a time T can be expressed as:

$$P_m(T) = \mathrm{Tr}\left(\rho \hat{N} \frac{(\alpha\hat{\theta})^m}{m!} e^{-\alpha\hat{\theta}}\right) \qquad (5.2.12)$$

where the operator N ensures that the subsequent operators are in normal order (see chapter 4).

5.3. Interpretation of the Photocounting Distribution Function

To interpret the expression for the photon-count distribution (eq. 5.2.7), we consider some simple examples. First, we will assume that the integrated intensity is independent of time, a case that occurs when the light comes from a perfectly stabilized laser, or when the coherence time of the light is much smaller than the measuring time T. Then we have

$$P(w) = \delta(w - w_0) \qquad (5.3.1)$$

and we get a Poisson distribution for the photon-counts:

$$P_m(T) = \frac{(\alpha w_0)^m}{m!} e^{-\alpha w_0} = \frac{(\langle m \rangle)^m}{m!} e^{-\langle m \rangle} . \qquad (5.3.2)$$

In the opposite case, where the measuring time T is much smaller than the coherence time τ_c of the light, we obtain from eq. 5.2.5

$$w = \int_t^{t+T} I(t'')dt' = TI(t) \qquad T \ll \tau_c \qquad (5.3.3)$$

and the photon-counting distribution becomes

$$P_n(T) = \int_0^\infty P(I) \frac{(\alpha TI)^m}{m!} e^{-\alpha TI} dI , \qquad (5.3.4)$$

where P(T) is the distribution function for the intensity. In the case of

of chaotic light, the distribution function of the fields is Gaussian and P(I) becomes an exponential distribution, i.e.

$$P(I) = \frac{1}{\langle I \rangle} e^{-I/\langle I \rangle} \tag{5.3.5}$$

and $P_m(T)$ becomes a Bose-Einstein distribution,

$$P_m(T) = \frac{\langle m \rangle^m}{(1+\langle m \rangle)^{m+1}} \tag{5.3.6}$$

where

$$\langle m \rangle = \alpha T \langle I \rangle .$$

For measuring times that are neither large nor small compared with the coherence time of the light, it is very difficult to find an explicit expression for the photon-counting distribution. For chaotic light with a Lorentzian spectral density, an expression for $P_m(T)$ has been found[27]. However, one can find an approximate expression for P_m with arbitrary measuring times T, a method which was first used by Rice (1945)[28] in connection with noise-theory. If we divide the counting time T into N parts, where each part is in the order of the coherence τ_c of the light, we can express the integrated intensity as a sum of independent terms, i.e.:

$$W = \int_t^{t-T} I(t')dt' = T/N \sum_{n=0}^{N} I(t+nT/N). \tag{5.3.7}$$

The generating function of the distribution function for the integrated intensity becomes:

$$F(s) = \int_0^{\infty} e^{-sw} P(w) = \langle e^{-sw} \rangle$$

and insertion of eq. 5.3.6 gives

$$\begin{aligned} F(s) &= \langle e^{-s\, T/N \sum I(t+nT/N)} \rangle \\ &= (\langle e^{-sT/N I(t)} \rangle)^N = (f(T/N\, s))^N \end{aligned} \tag{5.3.8}$$

where f is the generating function for the intensity.

To determine N we demand that our approximate distribution function has the proper second-order moment, i.e.,

$$\int_0^T \int_0^T I(t')I(t'')\rangle dt'dt' = T^2 \frac{N-1}{N}\langle I\rangle^2 + T^2 \frac{1}{N}\langle I^2\rangle \,. \qquad (5.3.9)$$

giving for the ratio T/N,

$$T/N = \frac{\frac{1}{T}\int_0^T \int_0^T \langle \Delta I(t')\Delta I(t'')\rangle dt'dt''}{\langle (\Delta I)^2\rangle} \,. \qquad (5.3.10)$$

which, as presumed, is of the order of the coherence time of the light. In the case of chaotic light, the distribution function for the intensity is as stated in eq. 5.3.5, and eq. 5.3.8 becomes in this case:

$$F(s) = \frac{1}{(1+s\frac{T\langle I\rangle}{N})^N} \,. \qquad (5.3.11)$$

By reversing eq. 5.3.11 we obtain the distribution function for the integrated intensity:

$$P(W) = \frac{N^N}{\langle w\rangle} \frac{(\frac{w}{\langle w\rangle})^{N-1}}{(N-1)!} e^{-N\frac{w}{\langle w\rangle}} \qquad (5.3.12)$$

and further for the photon-counting distribution we obtain

$$P_m(T) = \frac{\Gamma(N+m)}{m!\Gamma(N)} \frac{1}{(1+\alpha\frac{\langle w\rangle}{N})^N (1+N/\alpha\langle w\rangle)^m} \qquad (5.3.13)$$

where Γ is the gamma function.

By letting $N \to \infty$, which is equivalent to letting the coherence time τ_c be small compared with the counting time, eq. 5.3.13 become a Poisson distribution as it should be (eq. 5.3.2), and by letting $N \to 1$, eq. 5.3.12 becomes the Bose-Einstein distribution. In fig. 5.3 the photon-counting distribution is calculated for different values of the ratio T/τ_c.

Until now we have dealt with the problem of finding the distribution function for the counts of photo-electrons, with a known distribution function for the integrated intensity. The inverse problem of finding the probability function for the integrated intensities with a given photon-count distribution can in principle be solved. From the generating function for the integrated intensity given by

$$F(s) = \int_0^\infty P(w)e^{-sw}dw \qquad (5.3.14)$$

we can obtain the n^{th} order moments through the relation

$$\langle w^n \rangle = (-1)^n \frac{\delta^n F(s)}{\delta s^n}\Big|_{s=o}. \tag{5.3.15}$$

This means that the generating function can be expressed as:

$$F(s) = \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \langle w^n \rangle s^n. \tag{5.3.16}$$

From eq. 5.2.7 we then find

$$\langle w^n \rangle = \frac{1}{a^n} \cdot \langle m^{(n)} \rangle = \frac{1}{a^n} \langle m(m-1) \ldots\ldots (m-n) \rangle. \tag{5.3.17}$$

and by insertion of eq. 5.3.17 into eq. 5.3.16 we obtain

$$F(s) = \sum_{n=o}^{\infty} \frac{(-1)^n}{n!} \frac{1}{a^n} \langle m^{(n)} \rangle s^n. \tag{5.3.18}$$

By reversing eq. 5.3.18 we can then find P(w). From eq. 5.3.18 it can be seen that the generating function can be found if we know all the moments of the photon-count distribution. In practice, we only know the first few moments with sufficient accuracy, and the expansion in eq. 5.3.18 fails.

Another approach, that of Bedard[27], is to express the distribution function for the integrated intensity in terms of Laguerre polynomials,

$$P(w) = \sum_{p=o}^{\infty} a_p L_p(\alpha w) \tag{5.3.19}$$

where L_p is the Laguerre polynomial

$$L_p(x) = \sum_{q=o}^{p} \frac{(-1)^q}{q!} \binom{p}{q} x^q. \tag{5.3.20}$$

By using eqs. 5.2.7 and 5.3.19, one obtains

$$P(w) = \alpha \sum_{p} \Big(\sum_{q}^{p} (-1)^q \binom{p}{q} P_q(T) \Big) L_p(\alpha w). \tag{5.3.21}$$

This expansion gives a more accurate estimate of P(w) than eq. 5.3.18 for the reason that it suppresses the effect of the high counting rate with a small probability, which is not the case in eq. 5.3.18.

6. MEASUREMENT OF WIND VELOCITY IN THE ATMOSPHERE

A way to measure the wind velocity would be to use a hot-wire anemometer or a laser-doppler anemometer. Essentially these measurements would be point measurements; to obtain an estimate of the wind velocity over a larger volume, the measurement must be repeated several times at different space points, whereafter a suitable averaging procedure should be used. To overcome these difficulties, one could let the averaging be performed by a suitable optical configuration. Such an attempt has been reported recently[29] and commercial equipment has been developed. The optical configuration of this equipment is a transmitter and a receiver at a distance of 1 km from each other. A laser beam is transmitted from the transmitter to the receiver, which consists of two detectors placed around the optical axis at a distance of the order of the beam spot. The signals of the two detectors are correlated and a measure for the wind velocity is obtained. We will not further describe this equipment, but only mention that it is very complicated to calibrate. We propose another method to measure the wind velocity in the atmosphere.

The method in question is based on intensity correlation in a two-beam configuration (see fig. 6.1). The two laser beams are polarized perpendicular to each other and a polarization filter is placed in front of the detectors, so that the contribution to the intensity at one detector only originates from one laser beam. A disturbance moving with a velocity v in the x-direction (see fig. 6.1), giving a signal at a time t at detector 1, will give a signal at detector 2 at a time $t + \tau$. By correlating the intensity at the two detectors we can measure the time displacement τ, and then by knowing the distance between the detectors 2Δ, we can calculate the velocity as $2\Delta/\tau$.

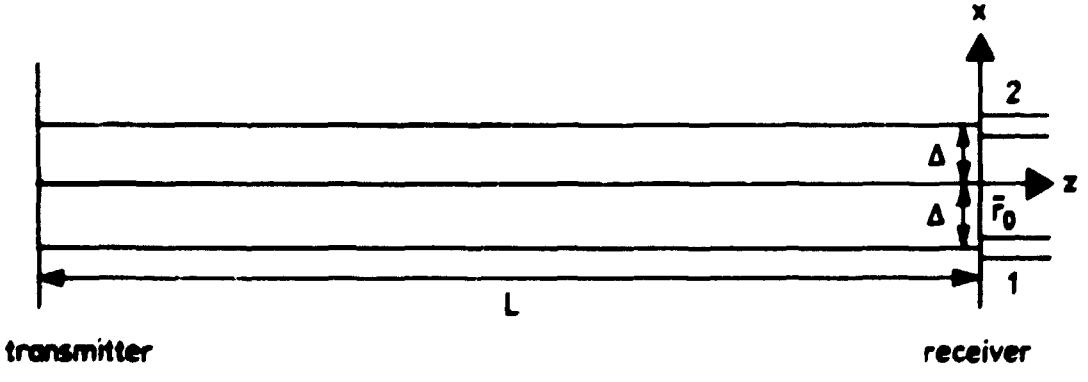


Fig. 6.1

Now, as mentioned, the idea of using this configuration was to perform an averaging in space, and the question is in which way do the different areas in space contribute to the intensity at the detector?. If we look at a disturbance at a distance z from the receiver where the laser beam width is w(z), a condition for a contribution at the detector must be that the correlation length l_c of the disturbance is greater than, or in the order of the beam spread w(z). This means that an area contributes only with disturbances in a certain range, and the whole configuration operates like a filter with respect to these correlation lengths. Another aspect of the arrangement is the distance between the two detectors 2Δ. By varying Δ at a time displacement of $\tau = 0$, we must expect to measure a quantity which is related to the correlation function for the disturbance. In the following analysis of the system it will appear that the covariance of the intensity is strongly related to the density correlation function for the medium; this is again related to the velocity correlation-function, which expresses the turbulence properties of the medium.

A way to describe the propagation of light through a random medium is to say that the medium has a fluctuating refractive index, which has a clear connection with geometric optics. This is the usual way however we will here describe the random properties by the density fluctuation of the medium. The reasons for this are that the density is a basic physical quantity in describing light scattering from a collection of particles, irregardless of whether the particles are molecules or are of macroscopic size, and the disturbance in the light passage through a medium is essentially a scattering process.

In order to analyze the system, we make certain assumptions about the properties of the medium. The medium is assumed to be stationary, homogeneous and locally isotropic. Assuming isotropicality in the medium as a whole would be an over-idealized assumption in the atmosphere, especially in the vertical direction.

In appendix B an expression is calculated for the electric field in a medium with a collection of scattering centres:

$$\bar{E}(r,t) = \bar{E}_o(r,t) + \int \bar{\bar{K}}(r-r,t-t') \cdot \bar{E}(r',t') \Delta n(r',t') dt' dr', \quad (6.1)$$

where $\bar{\bar{K}}(r-r', t-t')$ is a third-order tensor, $\Delta n(r', t')$ is the fluctuating part of the scattering particle density, and $\bar{E}_o(r, t)$ is the electric field in the undisturbed medium. The kernel function $\bar{\bar{K}}(r-r', t-t)$ has the form

$$\bar{\bar{K}}(r-r',t-t') = \frac{1}{8\pi^2\varepsilon_o} \int (\nabla\nabla + k^2\bar{\bar{I}}) \cdot \bar{\bar{\alpha}}(\omega) \frac{e^{ik|r-r'|}}{|r-r'|} e^{i\omega(t-t')} d\omega, \tag{6.2}$$

where $k = \omega/c$ and $\bar{\bar{\alpha}}(\omega)$ is the polarisability of the single scatter.

To simplify the expression for the kernel function, we can assume that the particle velocity is much less than the velocity of light and further that the polarizability is a scalar and a constant at the applied laser frequencies ω_o. The last assumption is equivalent to saying that the scatters are rotationally symmetric and that we ignore resonance phenomena.

$$\bar{\bar{K}}(r-r',t-t') = \frac{1}{4\pi\varepsilon_o} (\overline{\nabla\nabla} + k_o^2 \bar{\bar{I}}) \alpha(\omega_o) \frac{e^{ik|r-r'|}}{|r-r'|} \delta(t-t'). \tag{6.3}$$

The term $\overline{\nabla\nabla}$ in the kernel expresses the depolarized effect of the medium, and when, in this analysis of the configuration in fig. 6.1), we are only concerned with the forward scattering, we can neglect this term too. For the electric field, we then finally obtain:

$$\bar{E}(r,t) = \bar{E}_o(r,t) + \frac{k_o^2 \alpha(\omega_o)}{4\pi\varepsilon_o} \int \frac{e^{ik_o|r-r'|}}{|r-r'|} \bar{E}(r',t) \Delta n(r',t) dr'. \tag{6.4}$$

To solve this integral equation, we write the expression in the form

$$\bar{E} = \bar{E}_o + \hat{L}\bar{E}, \tag{6.5}$$

which can be solved to give

$$\bar{E} = \frac{1}{1-\hat{L}} \bar{E}_o = \sum_{n=0}^{\infty} \hat{L}^{(n)} E_o \tag{6.6}$$

where

$$\hat{L} = \frac{k_o^2 \alpha(\omega_o)}{4\pi\varepsilon_o} \int dr' \frac{e^{ik_o|r-r'|}}{|r-r'|} \Delta n(r',t).$$

The expression for the $\bar{E}$ field can be interpreted as follows:

$\bar{E}_o$ is the undisturbed field

$\hat{L}\bar{E}_o$ is the once scattered field,

$\hat{L}^2 E_o$ is the twice scattered field,

and so on.

Now if we let the light accepted by detectors 1 and 2 be polarized in the x and y directions respectively, we get for the intensities at detectors 1 and 2:

$$I_1(r_1) = \sum_{n,m} \hat{x}\cdot(\hat{L}_1^{(m)}\bar{E}_o)\ \hat{x}\cdot(\hat{L}_1^{(n)}\bar{E}_o)^* \tag{6.7}$$

and

$$I_2(r_2) = \sum_{n,m} \hat{y}\cdot(\hat{L}_2^{(m)}\bar{E}_o)\ \hat{y}\cdot(\hat{L}_2^{(n)}\bar{E}_o)^*. \tag{6.8}$$

By ensemble averaging we get, to the first order, the once scattered field:

$$\langle I_1(r_1)\rangle = \langle I_o(r_1)\rangle + \langle \hat{x}\cdot\hat{L}_1\bar{E}_o\ \hat{x}\cdot(\hat{L}_1\bar{E}_o)^*\rangle \tag{6.9}$$

and

$$\langle I_2(r_2)\rangle = \langle I_o(r_2)\rangle + \langle \hat{y}\cdot\hat{L}_2\bar{E}_o\ \hat{y}\cdot(\hat{L}_2\bar{E}_o)^*\rangle. \tag{6.10}$$

In the following we will assume that we can neglect the higher-order term in the expression for the E-field. This means that we limit the validity of the analysis to not too long a distance between the receiver and the transmitter, and to small turbulence intensity, or, to say it in another way, we must restrict ourselves to only consider areas where the field deviation from the undisturbed field is small. The available experimental data[31] indicate that this range is about 1 km in the atmosphere.

By correlating the fluctuating part of the intensities at the two detectors, we obtain by ensemble averaging:

$$\begin{aligned}&\langle (I_1(r_1,t) - \langle I_1(r_1)\rangle)\,(I_2(r_2,t+\tau) - \langle I_2(r_2)\rangle)\rangle \\ &\quad = \langle \Delta I_1(r_1,t)\,\Delta I_2(r_2,t+\tau)\rangle \qquad (6.11)\\ &\quad = \langle x\cdot E_o^*(r_1,t)\ y\cdot E_o(r_2,t+\tau)\ x\cdot(L_1E_o)\ y\cdot(L_2E_o)^*\rangle + \text{C.C.} \\ &\qquad \langle x\cdot E_o^*(r_1,t)\,y\cdot E_o^*(r_2,t+\tau)\,x\cdot(L_1E_o)\,y\cdot(L_2E_o)\rangle + \text{C.C.}\end{aligned}$$

Now, by setting $\bar{r}_1 = \bar{r}_o - \bar{\Delta}$ and $\bar{r}_2 = \bar{r}_o + \bar{\Delta}$, where $\bar{\Delta}$ is the distance between the detectors, we can express the undisturbed E-field as:

$$\bar{E}_o(\bar{r},t) = \hat{x}\, V_o(r-\bar{\Delta},t) + \hat{y}\, V_o(r+\bar{\Delta},t+\tau), \tag{6.12}$$

and we will assume the V_o is a Gaussian laser beam propagating in the z-direction, and with a beam waist w_o at $z = 0$,

$$V_o(r,t) = \frac{U_o \sigma}{\sigma + i k_o z}\, e^{-\frac{1}{2} k_o^2 \frac{x^2+y^2}{\sigma + i k_o z}}\, e^{i k_o z}\, e^{-i \omega_o t}$$

where

$$\sigma = 1/2\, k_o^2 w_o^2 .$$

Insertion of this in the correlation function gives:

$$\langle \Delta I(r,t)\ \Delta I(r_2,t+\tau)\rangle = \frac{|\alpha(\omega_o)|^2\, k_o^4}{(4\pi\epsilon_o)^2}\, |V(r_o)|^2 \times$$

$$\iint \frac{e^{-ik_o|\bar{r}_o-\bar{r}_1'|}}{|r_o-r_1'|}\, \frac{e^{ik_o|\bar{r}_o-\bar{r}_2'|}}{|r_o-r_2'|}\, V_o^*(\bar{r}_1')\, V_o(\bar{r}_2') \tag{6.13}$$

$$\langle \Delta n(\bar{r}_1'+\bar{\Delta},t)\ \Delta n(\bar{r}_2'-\bar{\Delta},t+\tau)\rangle\, dr_1'\, dr_2' + \text{C.C.}$$

$$+ V(r_o)^2 \iint \frac{e^{-ik_o|\bar{r}_o-\bar{r}_1'|}}{|r_o-r_1'|}\, \frac{e^{ik_o|\bar{r}_o-\bar{r}_2'|}}{|r_o-r_2'|}\, V_o(\bar{r}_1')\, V_o(\bar{r}_2')$$

$$\langle \Delta n(\bar{r}_1'+\bar{\Delta},t)\ \Delta n(\bar{r}_2'-\bar{\Delta},t+\tau)\rangle\, dr_1'\, dr_2' + \text{C.C.}\Big\}$$

As previously assumed, the density fluctuation is stationary and locally isotropic. The assumption of local isotropicality means that the covariance function for the density in two space points, $\bar{r}_1$ and $\bar{r}_2$, will vary rapidly as a function of the distance $\bar{r}_2-\bar{r}_1$ and vary slowly with mean position $\frac{1}{2}(r_1+r_2)$. With this in mind we can write the density covariance function as:

$$\langle \Delta n(\bar{r}_1'+\bar{\Delta},t)\ \Delta n(\bar{r}_2'-\bar{\Delta},t+\tau)\rangle = F(\tfrac{1}{2}(\bar{r}_1'+\bar{r}_2'),\ \bar{r}_2'-\bar{r}_1'-2\bar{\Delta},\tau).$$

By inserting the Fourier transform of F with respect to $\bar{r}'_2-\bar{r}'_1$, we obtain the following expression for the intensity covariance function

$$\langle \Delta I(r,t)\ \Delta I(r_2,t+\tau)\rangle = \frac{n_o^2|\alpha(\omega_o)|^2 k_o^4}{(4\pi\varepsilon_o)^2}\,|V(r_o)|^2 \times$$

$$\frac{1}{(2\pi)^3}\int F(\tfrac{1}{2}(\bar{r}'_1+\bar{r}'_2),k,\tau)\ e^{-i\bar{k}\cdot 2\Delta}\ H(k,\bar{r}'_1,\bar{r}'_2)\ d^3r'_1\ d^3r'_2\ d^3k \tag{6.14}$$

where

$$H(k,r'_1,r'_2)=\left\{\frac{e^{ik_o|\bar{r}_o-\bar{r}'_2|-ik_o|\bar{r}_o-\bar{r}'_1|}}{|r_o-r'_1|\ |r_o-r'_2|}\,V_o^*(\bar{r}'_1)V_o(\bar{r}'_2)+\text{C.C.}\right\}e^{i\bar{k}\cdot(\bar{r}'_1-\bar{r}'_2)}$$

$$+\frac{V_o^*(r_o)}{V_o(r_o)}\left\{\frac{e^{ik_o|\bar{r}_o-\bar{r}'_2|+ik_o|\bar{r}_o-\bar{r}'_1|}}{|\bar{r}_o-\bar{r}'_2|\,|\bar{r}_o-\bar{r}'_1|}\,V_o(r'_1)V_o(r'_2)+\text{C.C.}\right\}e^{i\bar{k}\cdot(\bar{r}'_1-\bar{r}'_2)}$$

Now, by expanding the square term in the expression for the function $H(k,\bar{r}'_1,\bar{r}'_2)$ and by setting $1/2(\bar{r}'_1+r'_2 = 1/2(z'_1+z'_2) = z$, we can perform the integration in the x and y directions, and finally express the intensity covariance function as

$$\langle \Delta I(r,t)\ \Delta I(r_2,t+\tau)\rangle = \frac{n_o^2|\alpha(\omega_o)|^2 k_o^2}{4\Sigma_o^2}\,|V(z_o)|^4 \times \tag{6.15}$$

$$\frac{1}{(2\pi)^3}\int F(z,k,\tau)\ e^{-ik\cdot 2\Delta}\ G(k,z)\ dz\ d^3k$$

where

$$G(k,z)=\int_{2(z_o-z-L)}^{\quad} e^{-\frac{\sigma k_t^2}{\sigma^2+k_o^2z_o^2}(z_o-z)^2+\frac{i}{4}\rho^2)}$$

$$\times\ e^{-i\left(1-\frac{2k_oz_o(z_o-z)}{\sigma^2+k_o^2z_o^2}\right)\frac{k_t^2}{2k_o}}-e^{-i\left(\frac{z_o-z}{k_o}-k_oz_o\frac{(z_o-z)^2+\frac{i}{4}\rho^2}{\sigma^2+k_o^2z_o^2}\right)k_t^2}\Big]e^{ik_z}\ d \tag{6.16}$$

and z_o is the position of the receiver plane relative to the beam waist of the laser beam, L the distance between the transmitter and the receiver, and k_t the transverse wave number.

With this expression we have adopted a mode-description of the scattering process. The function G(k, z) serves as a filter function, determined by the Gaussian wave form and the length between the transmitter plane and the receiver plane, and it describes to what extent a mode at a position z along the optical axis will contribute to the intensity fluctuation at the receiver. In general, the expression for the weight function G(k, z) is complicated and a calculation must be performed numerically.

The dependence of the wave number in the z-direction, k_z, is very rapid and has the character of a delta function, a property which can be expected for physical reasons. Modes in the z-direction with a wavelength smaller than the distance between the transmitter and the receiver will be averaged out.

For small wavelength F(z, k, τ) is slowly varying, so by setting

$$F(z,k,\tau) = F(z,k_t,o,\tau) = F_1(z,k_t,\tau)$$

we obtain by performing the integration in the k_z direction:

$$\langle \Delta I(\bar{r}_1,t)\Delta I(\bar{r}_2,t+\tau)\rangle = \frac{n_o^2|\alpha(\omega_o)|^2k_o^2}{(4\pi\varepsilon_o)^2}|V(z_o)|^4$$

$$\times\frac{1}{(2\pi)^2}\int F_1(z,k_t,\tau)e^{-ik_t^2\Delta}G_1(k_t,z)dz\,d^2k_t + C.C.$$

where

$$G_1(k_t,z) = \int G(k,z)dk_z$$

$$= e^{-\frac{\sigma k_t^2(z_o-z)^2}{\sigma^2+k_o^2z_o^2}}\left\{1 - e^{-i\frac{z_o-z}{k_o}\frac{\sigma^2+k_o^2z_oz}{\sigma^2+k_o^2z_o^2}k_t^2}\right\}$$

From eq. 6.18 it is seen that $G_1(k_t, z)$ only allowed modes with the wavenumbers in the range of

$$k = p\frac{\pi}{2}\left(\frac{k_o(o^2+k_o^2z_o^2)}{(z_o-z)(o^2+k_o^2z_oz)}\right)$$

to give contributions to the intensity covariance function.

Now, by assuming that the variations of the density correlation function with respect to the time displacement τ can be expressed by a displacement in space by the amount $v(z)\tau$, where $v(z)$ is the velocity of the medium at the position z, we obtain for the intensity covariance function:

$$\langle \Delta I(r,t)\Delta I(r,t+\tau)\rangle = \frac{n_o^2|\alpha(\omega_o)|^2k_o^2}{(4\pi\epsilon_o)^2}|V_o(z_o)|^4 \qquad (6.19)$$

$$\times \frac{1}{\pi}\int kF_2(z,k)J_o(k|2\Delta - v(z)\tau|)G_2(k,z)dzdk$$

where J_o is the Bessel function of ordre zero, G_2 is the real part of G_1 and $F_2(z,k)$ the turbulence spectrum of the atmosphere, which have the functional form[30]:

$$F_2(z,k) = A(z)\,k^{-11/3}\,e^{-k^2/k_m^2} \qquad (6.20)$$

where k_m gives the inner scale of turbulence in the atmosphere, and $A(z)$ is a structure factor which depend on $z \cdot k_m$ is ordinary in the ordre of $10\ mm^{-1}$ [30].

Now our aim was to measure the wind velocity in the atmosphere. If the cross-wind speed was constant between the transmitter and the receiver plane, it is clear that the intensity covariance function would have a maximum for the time lag $\tau = 2\Delta/v$. If the spread in the cross-wind speed is large, then the intensity covariance function would be smeared out, and a measurement in this case would be meaningless with this equipment. By assuming that the variation in the wind speed is small, we can develop the intensity covariance function about its maximum with respect to the time lagτ. The time varying part can be expressed by:

$$-(2\Delta - v(z)\tau)^2\,w(z)dz \qquad (6.21)$$

where $w(z)$ gives the weights for the velocity along the z-axis. $w(z)$ is given by:

$$w(z) = \int k^3 F_2(z,k) G_2(z,k) dk \qquad (6.22)$$

From eq. 6.21 we find that the mean cross-wind speed v in areas where w(z) gives a uniform weighting is given by:

$$\langle v \rangle = \tfrac{1}{2}(v_o + \sqrt{v_o^2 + \langle (v - \langle v \rangle)^2 \rangle}) \qquad (6.23)$$

$$v_o = 2\Delta/\tau_o$$

where τ_o is the time lag at which the intensity covariance function has its maximum value. $\langle (v - \langle v \rangle)^2 \rangle$ is the variance of the cross-wind speed.

The weight function w(z) depend on the structure factor A(z), the path length L and the magnitude and position of the beam waist of the laser beam. In fig. 6.2 we have sketch w(z) for different path lengths, with beam waist placed at the transmitter plane and with a beam waist of zero (spherical

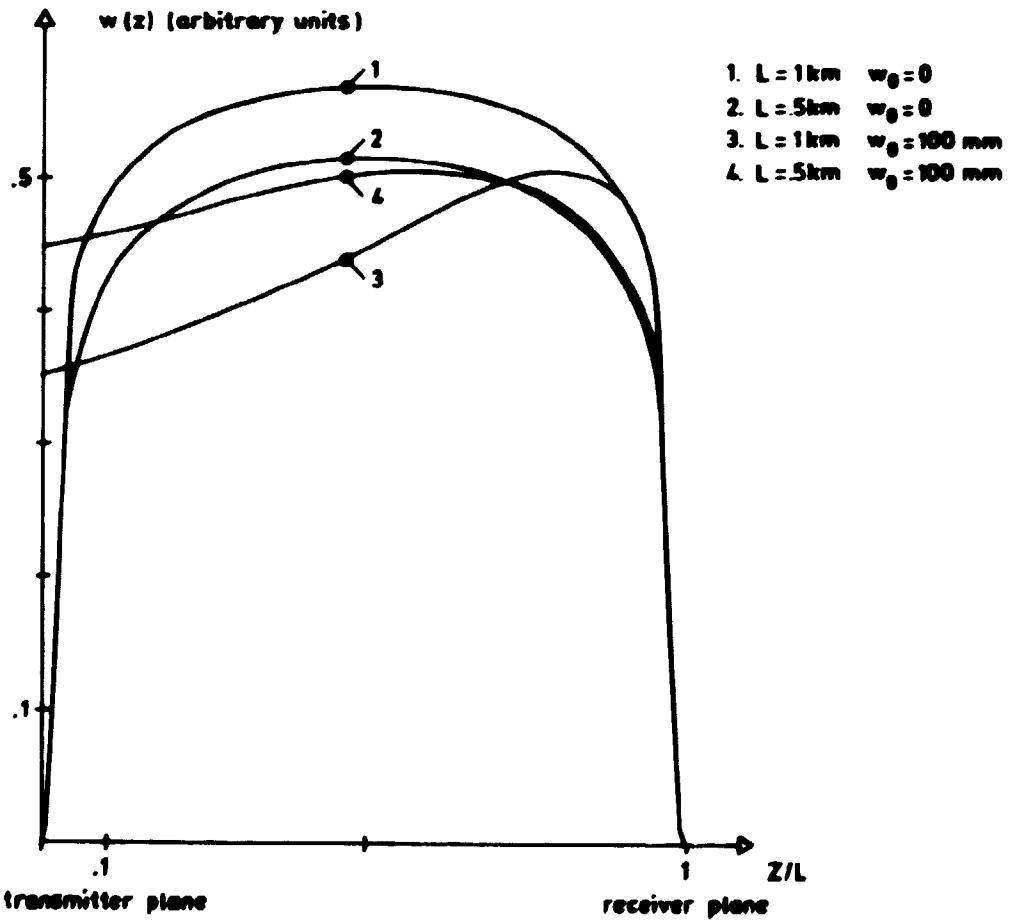


Fig. 6.2

wave) and of 100 mm. The inner scale l_o of turbulence is assumed to be 1 mm ($k_m = 2\pi/l_o$), and the structure factor is assumed to be constant.

From fig. 6.2 it is seen that care must be take if we want a uniform weighting function. The condition for a uniform weighting function can be expressed by the condition:

$$w_o^2(L) \qquad \frac{1}{\pi}\left(\frac{Lk_m}{k_o}\right)^2 \tag{6.24}$$

where $w_o(L)$ is the beam width of the laser at the receiver plane.

7. SINGLE-BURST DETECTION IN A LASER-DOPPLER VELOCITIMETER SYSTEM

This part of the report describes the detection of a transient signal that is modulated through a stochastic process. The idea is to analyze a specific detection system[32], or rather an idealization of this system, in order to optimize the amount of information obtainable from the detection procedure. To do this, we first give a brief description of a Laser-Doppler velocitimeter system and define some characteristic parameters for the system.

By crossing two laser beams we get an interference pattern at the intersection point. When a particle passes the interference pattern, part of the light will be scattered. The intensity of the scattered light is modulated with the Doppler frequency[33, 34],

$$\omega_o = \Delta k \cdot v, \tag{7.1}$$

where Δk is the difference between the wave vectors of the two laser beams.

The intensity of the scattered light, the Doppler signal, can be expressed by

$$I(t) = I_o e^{-t^2/\tau_c^2} (1 + \gamma \cos \omega_o t), \tag{7.2}$$

where the Gaussian amplitude factor expresses the fact that the intensity of light in the measuring volume has a Gaussian distribution. τ_c is the running time of the particle through the measuring volume, and it can be related to the Doppler frequency and the number of interference lines by

$$\tau_c = n/\omega_D. \tag{7.3}$$

The detection process is carried out by a photomultiplier followed by a photon counter and a correlator that autocorrelates the signal at a time T, the measuring time. The autocorrelation function is then Fourier-transformed, and we get an estimate of the power spectrum of the incoming light as the output. The photon counter counts the number of photo electrons in a time increment Δt, so the autocorrelation function is a correlation between counts. The detection procedure is sketched in fig. 7.1.

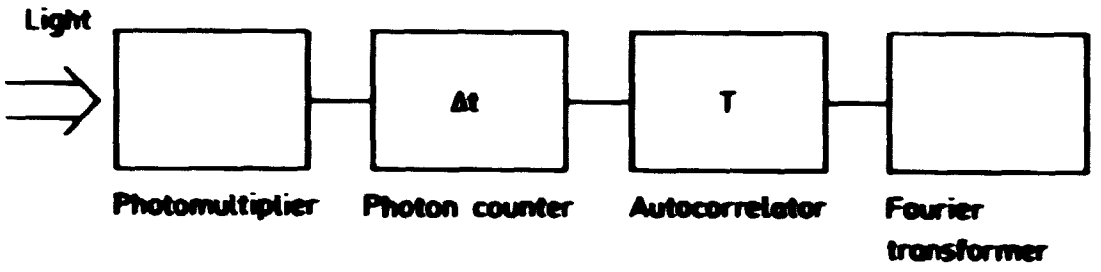


Fig. 7.1

The incoming light of the photomultiplier is essentially a stochastic signal, but if we choose the counting time of the photon counter Δt so that it is greater than the coherence time of the laser, then we can consider the scattered light as a deterministic signal. The counting time Δt is also restricted by the Doppler frequency; if we are to resolve a periodical signal with a frequency ω_D, we must choose Δt as:

$$\Delta t \ll 1/\omega_D.$$

For Doppler frequencies up to 1 Mhz, the counting time Δt can be chosen so that it is much greater than the coherence time of laser light and this is assumed throughout the analysis. Another aspect of this assumption is that counts in different time intervals will be independent in a statistical sense.

The counts from the photon counter will have a Poisson distribution[35] due to the discrete nature of the photodetecting process; the probability to count n photoelectrons at a time interval $(t/t+\Delta t)$ is then:

$$p_n(t \quad t+\Delta t) = \frac{\langle n(t) \rangle^n}{n!} e^{-\langle n(t) \rangle}, \qquad (7.4)$$

where $\langle n(t) \rangle$ is the mean counts given by

$$\langle n(t) \rangle = I(t)\,\Delta t, \tag{7.5}$$

and I(t) is the current of the incoming light. If the number of counts is great ( )100) when the particle is in the measuring volume, the fluctuations in the number of counts will be negligible and the power spectrum of the counts will represent the "true" spectrum of the scattered light as in an actual deterministic case. The problem arises, however, when we are concerned about the number of counts of the order of one, a situation which is realistic in a case where the detector equipment is located far from the measuring volume. The fluctuations in the number of counts will cause the spectrum to deviate from the "true" spectrum and the Doppler peak in the spectrum will be difficult to identify.

If we let i(t) be the current from the photon counter at the time t, we get for the autocorrelation function:

$$R(\tau) = \int_{-T/2}^{T/2} i(t)\; i(t+\tau)\; dt, \tag{7.6}$$

where T is the measuring time.

The Fourier component of i(t) is:

$$S_q = \int_{-T/2}^{T/2} i(t)\; e^{-i\frac{2\pi}{T} qt}\; dt \tag{7.7}$$

and the Fourier transform of the autocorrelation function R( ) can be expressed by:

$$R_q = |S_q|^2 . \tag{7.8}$$

This means that the power spectrum of the current is equal to the absolute square of the amplitude spectrum of the currents, and we can analyse the power spectrum by analysing the amplitude spectrum. The current i(t) is a constant in the time intervals $(t_p - 1/2\,\Delta t,\ t_p + 1/2\,\Delta t)$, and the amplitude spectrum of the current can be written as

$$
\begin{aligned}
S_q &= \int_{-T/2}^{T/2} i(t)\, e^{-i\frac{2\pi}{T}qt}\, dt \\
&= \sum_p i(t_p)\, \frac{1}{T} \int_{t_p-\frac{1}{2}\Delta t}^{t_p+\frac{1}{2}\Delta t} e^{-i\frac{2\pi}{T}qt}\, dt \\
&= \sum_p n(t_p)\, e^{-i\frac{2\pi}{T}qt_p}\, \left(\frac{\sin\frac{\pi}{T}\Delta tq}{\pi/T\ \Delta tq}\right)
\end{aligned}
\tag{7.9}
$$

where $n(t_p)=i(t_p)\Delta t$, and Δt is the number of counts in the time intervals $(t_p - 1/2\,\Delta t,\ t_p + 1/2\,\Delta t)$. The sin x/x factor reflects the fact that no frequency above $\pi/2\Delta t$ will be present in the spectrum, and we will, for simplicity, neglect this factor by restricting ourselves to only consider frequency components that fulfil the condition:

$$q < T/\Delta t = N,$$

where N is the number of samples.

So we can write:

$$S_q = \sum_p n(t_p)\, e^{-i\frac{2\pi}{T}qt_p}\,. \tag{7.10}$$

By ensemble averaging we get for the mean spectrum

$$\langle S_q \rangle = \sum_p \langle n(t_p) \rangle\, e^{-i\frac{2\pi}{T}qt_p} \tag{7.11}$$

and for the variance of the spectrum:

$$
\begin{aligned}
V(S_q) &= \sum_{p,l} \left(\langle n(t_p)\ n(t_l)\rangle - \langle n(t_p)\rangle\langle n(t_l)\rangle\, e^{-i\frac{2\pi}{T}q(p-l)}\right. \\
&= \sum_p \langle n(t_p)\rangle\,.
\end{aligned}
\tag{7.12}
$$

The last expression is obtained by using the properties of the Poisson statistics.

The mean number of counts at the time t_p is given by the Doppler signal:

$$\langle n(t_p)\rangle = \Delta t\ I_o\, e^{-t_p^2/\tau_c^2}\, (1 + \gamma \cos\omega_D\, t_p). \tag{7.13}$$

Now, the value of the current I_o depends on several parameters. It is proportional to the intensity in the measuring volume, the area of detection, the quantum efficiency of the photo tube, and it is inversely proportional to the square of the distance to the measuring volume. We are here only interested in the dependence of the number of interference lines in the measuring volume, so we can write (the Doppler frequency is assumed constant):

$$I_o = K_o/n^2 . \tag{7.14}$$

The mean spectrum and the square roots of the variance have been calculated for different values of the number of interference lines and the result is sketched in fig. 7.2. The time scaling in the calculation is set so that the product of the Doppler frequency and the time increment is a unit, and the measuring time T is:

$$T = N\,\Delta t ; \quad N = 63.$$

This scaling implies that the Doppler frequency will lie in the spectrum at mode number 10. From the calculated spectra we see that the Doppler frequency represents a peak in the spectra, and the relative spread of the spectra increases with increasing values of the number of interference lines, or in usual electronic terms we could say that the signal to noise ratio decreases with an increasing number of interference lines. Generally, we wish to maximize the signal to noise ratio, but in this case it would mean that the number of interference lines should be small, which would cause the spectrum to be very broad and it would be very difficult to identidy the Doppler frequency in an actual spectrum. This fact implies a value of the number of interference lines that optimizes the properties of the detection system. In order to find such optimum values, which maximize the visibility of the Doppler frequency in the spectrum, we will compare the frequency spread in the Doppler signal with the spectral resolution. The frequency spread in the Doppler signal is:

$$(\Delta\omega)_D = \sqrt{2}/\tau_c$$

and the spectral resolution is:

$$\Delta\omega = 2\pi/T .$$

The spectral resolution gives the maximum accuracy by means of which we can determine the Doppler frequency, and it will be of no use to let the frequency spread in the Doppler signal be much smaller than the spectral

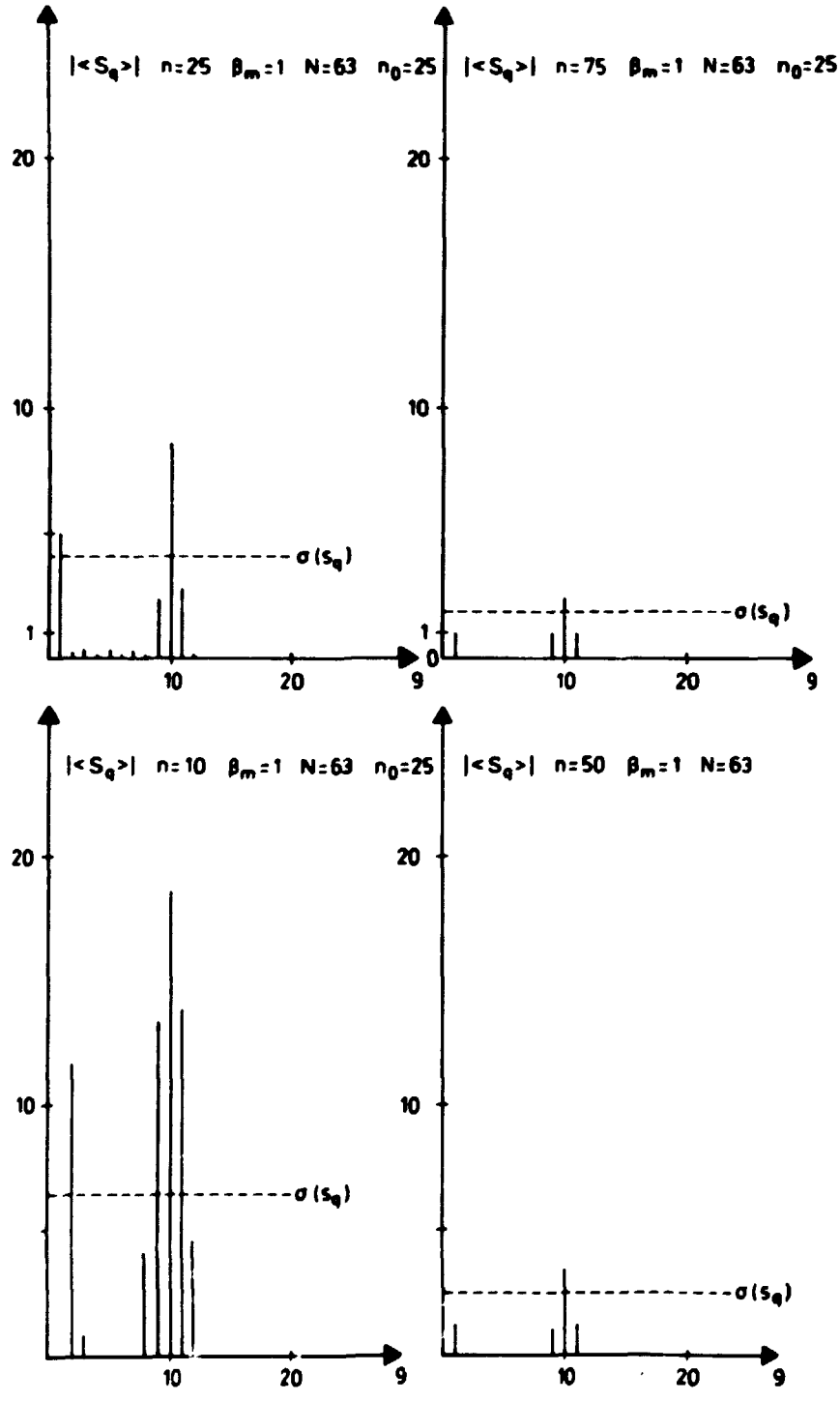

|<S_q>| n=25 β_m=1 N=63 n_0=25
|<S_q>| n=75 β_m=1 N=63 n_0=25
|<S_q>| n=10 β_m=1 N=63 n_0=25
|<S_q>| n=50 β_m=1 N=63
σ(s_q)
20
10
1
0
9

Fig. 7.2

resolution in order to improve this accuracy. Further, it would give more "noise" in the system as stated above. On the other hand, if we let the frequency spread in the Doppler signal be greater than the spectral resolution, it would be this quantity which would determine the accuracy from which we could determine the Doppler frequency, and it would be worse than the accuracy obtained in the first case. The best case, which maximizes the signal to noise ratio and simultaneously gives the least uncertainty in the determination of the Doppler frequency, can be estimated to be:

$$(\Delta\omega)_D \approx 1/2\,(\Delta\omega)\,.$$

The factor 1/2 implies that the neighbour modes to the Doppler mode will be suppressed.

From the above expression we get:

$$\tau_c \approx \frac{\sqrt{2}}{\pi} T\,,$$

or, by using $\omega_D \cdot \Delta t = 1$,

$$n = \frac{\sqrt{2}}{\pi} N,$$

which, for the spectra sketched, fig. 7.2, gives an optimum value of 28 for the number of interference lines.

Now, to give this verbal description a more concrete form, we will define a parameter β, which can give the optimum value for the number of interference lines, and, as we shall see, also give a qualitative measure for the figure of merit of the detection system. The parameter β is defined as the ratio of the distance between the Doppler mode and its neighbour mode in the mean spectrum and the spread in the spectrum at the Doppler mode:

$$\beta = \frac{\langle S_{q_o} \rangle - \langle S_{q_o+1} \rangle}{\sqrt{V(S_{q_o})}}$$

where q_o is the Doppler frequency mode number. The parameter β is a function of the number of interference lines. A high value of β means that the Doppler peak in the spectrum is clearly visible, so to get a "efficient" detection system we must maximize β. Using statistical language, we could say that β describes the degree of overlap for the distribution function for the Doppler mode and its neighbour modes. If the fluctuation part of the modes in the spectrum has a Gaussian distribution, then a β value of six for the system would mean that the frequency peak

in an actual spectrum would be the Doppler frequency with a probability of 99.9%. In this way, we could say that the parameter β describes the degree of determinism in the detection system.

According to the earlier discussion, we must expect that β as a function of the number of interference lines n has a maximum value, and that it goes to zero for n equal to zero and for n going to infinity. With the time scaling as before, we have calculated β/β_{max} as a function of n. The result is sketched in fig. 7.3. The maximum value of β is achieved for a value of the number of interference lines obtained above (n = 28). The maximum value of β, β_m, is related to the quantity K_o through

$$\beta_m = \frac{1}{n_o} \beta_o \sqrt{K_o \Delta t}$$

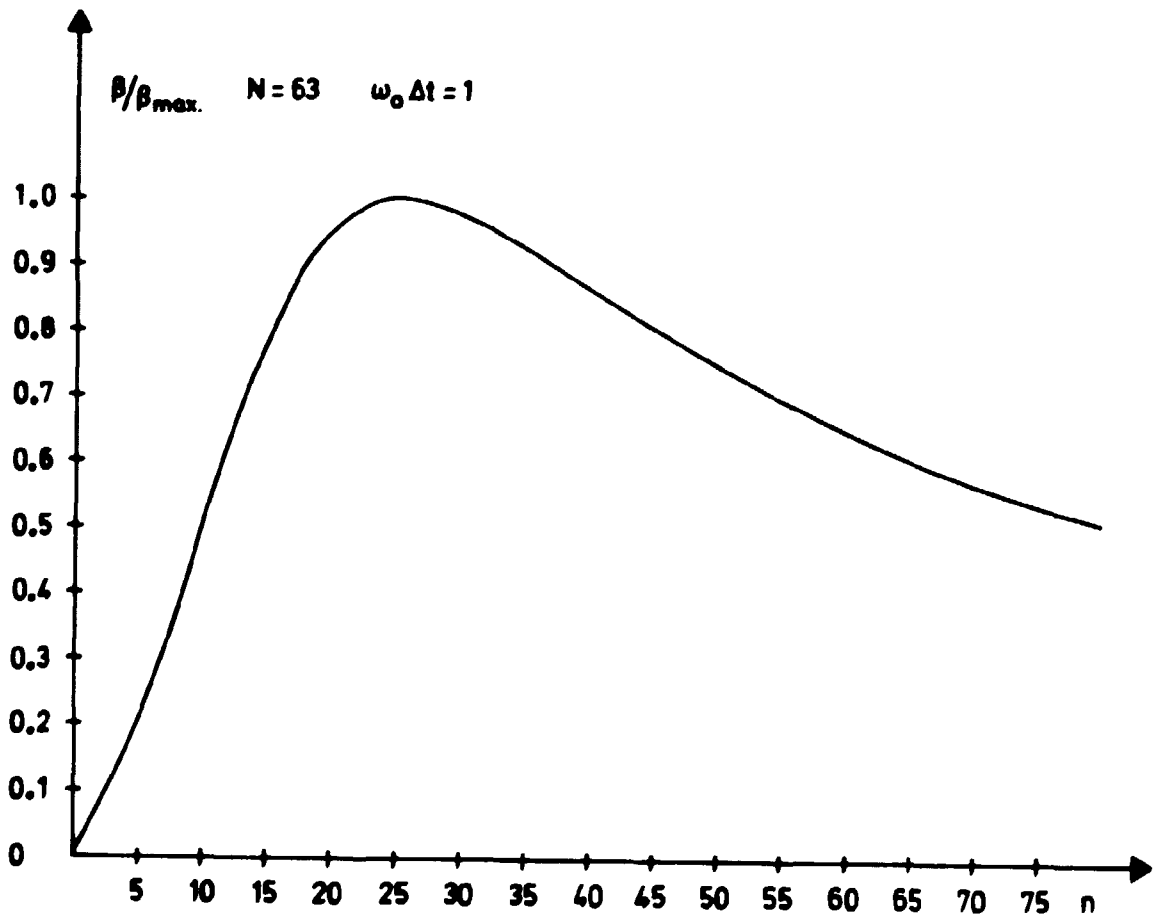


Fig. 7.3

where β_o is in this case calculated to 1.105.

From this expression we can write the mean number of counts at the time t_p as:

$$\langle n(t_p) \rangle = \left(\frac{n_o}{n}\right)^2 \left(\frac{\beta_m}{\beta_o}\right)^2 e^{-t^2/\tau_c^2} (1 + \gamma \cos\omega_D t_p). \qquad (6.16)$$

All the curves shown are calculated with a β_m and a γ value of one.

To give an impression of the actual spectra and of the effect of various numbers of interference lines, we simulated the spectra on a programmable pocket calculator. The two sets of curves sketched in figs. 7.4 and 7.5 are obtained by two different sets of "randomness", and with the same parameters as in the curves for the mean spectra. From the simulated spectra, we see that the Doppler frequency is clearly visible in the spectra with the optimum values of n, whereas the other value of n gives a poorer picture.

Conclusion

Hitherto we have neglected the noise contribution from the background and from small particles running through the measuring volume. This kind of noise will increase the fluctuation part of the spectra and we must expect the optimum value of n to decrease, and with it the maximum obtainable β value. However, in spite of the noise, the analysis given here can serve as background for a real experiment. The number of interference lines should be kept below the optimum value given by this kind of analysis, and we may also say something about the range of the detecting system, as the maximum value of β is inversely proportional to the distance between the detector and the measuring volume. At a certain distance, if we obtain a β value of 2, i.e. the system is almost "deterministic" and we can make instantaneous velocity measurements, an increase of the distance by a factor 10 would give a β value of 0.2, and an additional averaging must be made to obtain the correct wind velocity.

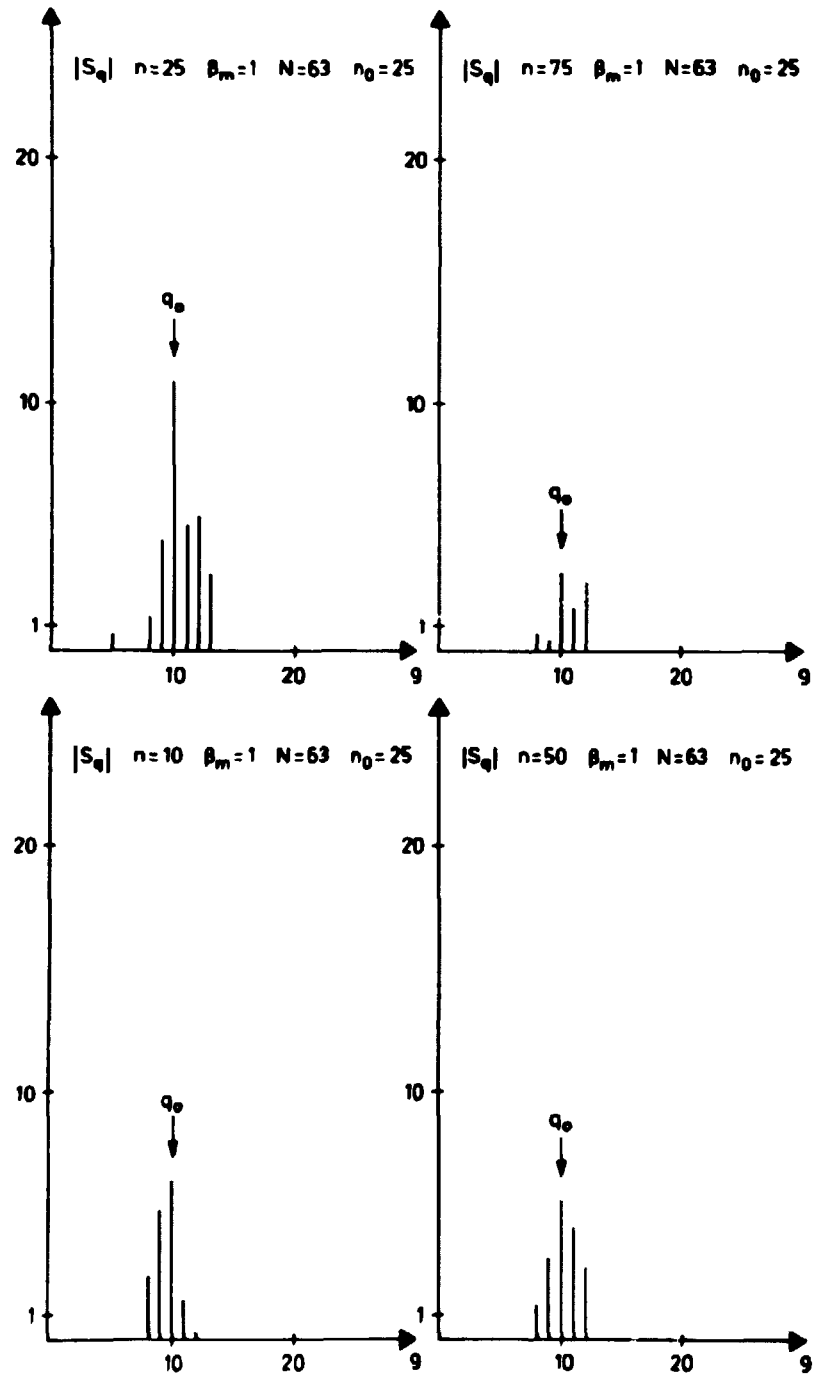

|S_q| n=25 β_m=1 N=63 n_0=25
|S_q| n=75 β_m=1 N=63 n_0=25
|S_q| n=10 β_m=1 N=63 n_0=25
|S_q| n=50 β_m=1 N=63 n_0=25
q_e
20
10
1
10
20
9

Fig. 7.4

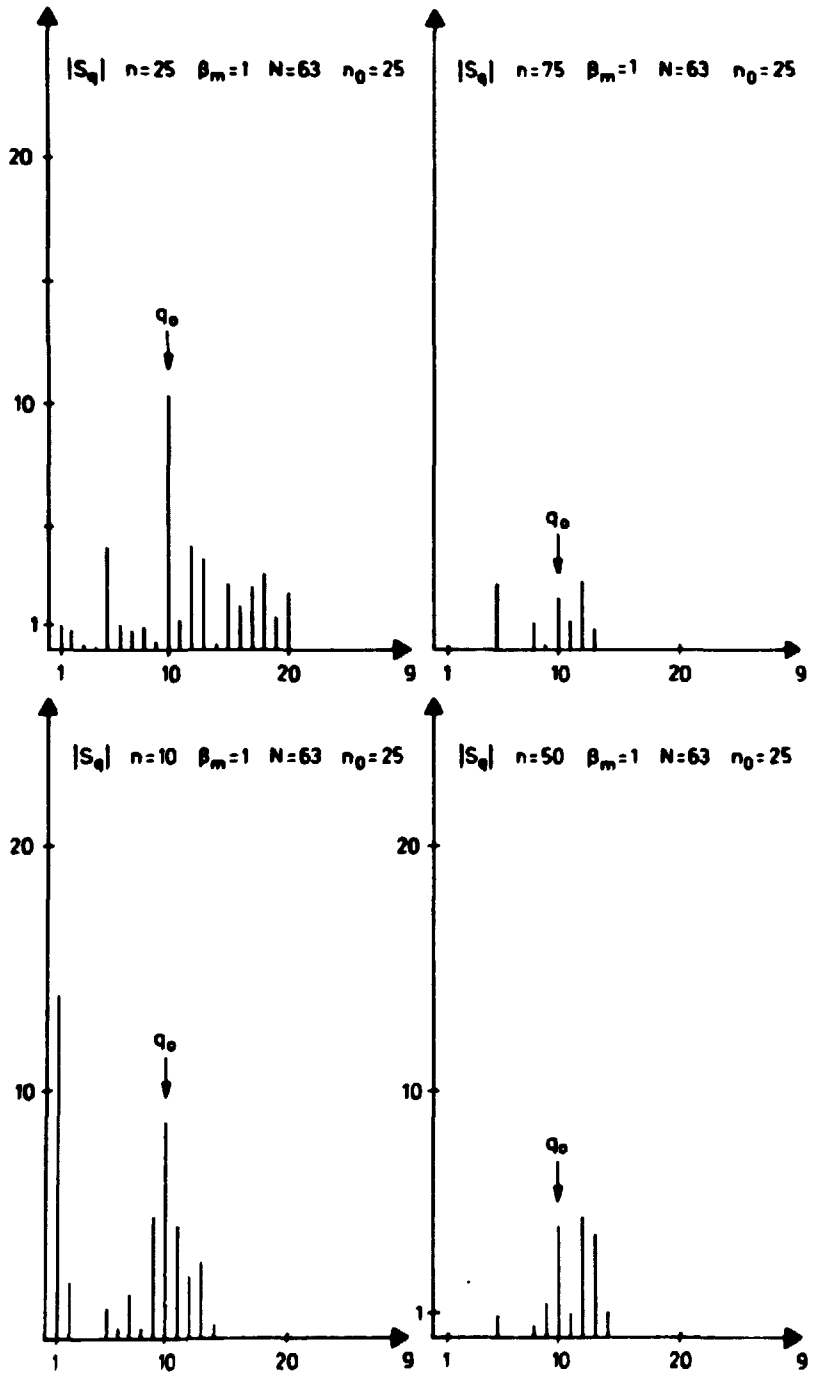

|Sq| n=25 βm=1 N=63 n0=25
|Sq| n=75 βm=1 N=63 n0=25
|Sq| n=10 βm=1 N=63 n0=25
|Sq| n=50 βm=1 N=63 n0=25
q0
20
10
1
1
10
20
9

Fig. 7.5

8. LIGHT SCATTERING FROM A COLLECTION OF INDEPENDENT SCATTERING CENTRES

8.1. The Intensity-Intensity Correlation Function

In this section we wish to investigate the information obtainable from a light scattering experiment with independent scatterers by measuring the second- and the fourth-order correlation function.

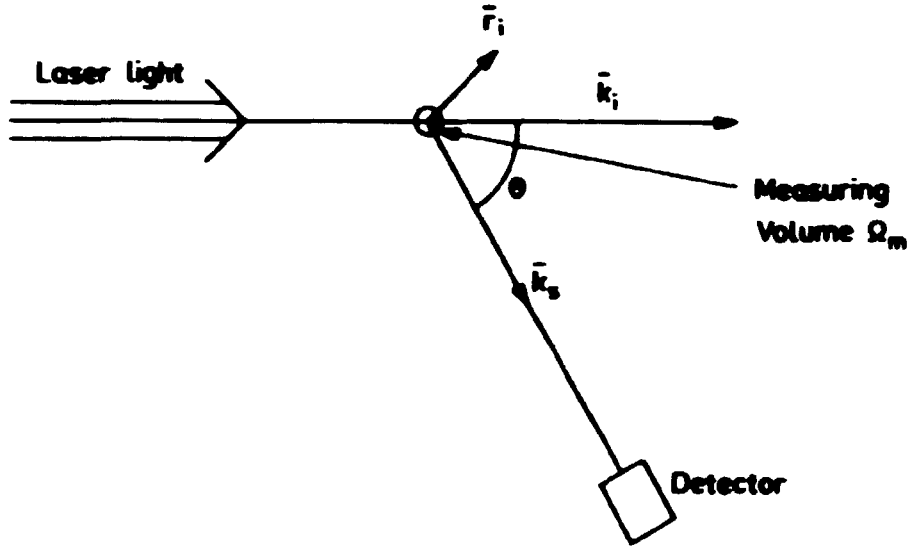


Fig. 8.1

The scattered light is collected from an area of finite extent, and it is limited by the optical equipment, by the intensity distribution of the light source, or both.

The second-order correlation function is obtained by using heterodyne technique, i.e. the photodetector is simultaneously illuminated by the scattered light and the laser light transmitted directly in the detector, which acts as a local oscillator signal. The fourth-order correlation function can be measured by using self-beating. The correlation functions calculated here are calculted on one point on the photodetector surface, so the results obtained are only valid when we deal with coherent detection(8.2).

The statistical properties of the scattered field will depend on the light source and the shape and motion of the scattering particle, and furthermore on the finite extent of the measuring volume. We will assume that the scattering particle is of macroscopic size and, for simplicity, of a spherical

shape. The particles are embedded in a medium which supports them without giving any essential contribution to the scattered field, and we will assume that the scattering particles move without any mutual interaction, so their motions are determined by the movement of the surrounding medium and the individual motions of particles. By using a laser as light source, we can consider the scattered field as a deterministic signal modulated by an indeterministic process, determined by the motion of the scattering particles. In a case where the properties of the medium are deterministic and the mean number of the scattering particles in the measuring volume is "great", so that we can use the central limit theorem for the total scattered field, the statistics of the field are Gaussian and all the information is contained in the second-order correlation function of the field. For instance, the fourth-order correlation function is given by the Siegert relation:

$$g^{(4)}(t,\tau) = 1 + |\, g^{(2)}(t,\tau)\,|^2 .$$

If the properties of the medium are indeterministic, the statistics of the scattered field will not in general be Gaussian, but formally we can use the properties of the field with Gaussian statistics to calculate the fourth-order correlation function. If $g^{(2)}(t,\tau)$ in the above expression is the result of an ensemble-averaging over the initial position of the scattering particles, then the fourth-order correlation function is obtained by an ensemble averaging of the Siegert relation over the stochastic parameter related to the medium. Using the letters P and M to denote, respectively, ensemble-averaging over the intial position of the particle and the stochastic parameters of the medium, we get:

$$\langle g^{(4)}(t,\tau)\rangle_P = 1 + |\,\langle g^{(2)}(t,\tau)\rangle_P\,|^2$$

and

$$\langle\langle g^{(4)}(t,\tau)\rangle_P\rangle_M = 1 + \langle|\langle g^{(2)}(t,\tau)\rangle_P|^2\rangle_M \neq 1 + |\langle\langle g^{(2)}(t,\tau)\rangle_P\rangle_M|^2 .$$

In the following we will calculate the second- and fourth-order correlation functions with an arbitrary number of particles in the measuring volume by averaging over the initial particle position, whereafter two cases are considered; one where the particles undergo Brownian motion, the other an example of a turbulent medium.

The scattered electrical field is a sum of the field scattered from the single particles, and it is written

$$E_s(t) = \sum_i E_s(\bar{r}_i(t)), \tag{8.1.1}$$

where $\bar{r}_i(t)$ is the position of the i^{th} particle.

$E_s(\bar{r}_i(t))$ can be expressed as (see fig. 9.1):

$$E_s(\bar{r}_i(t)) = I_o^{\frac{1}{2}} P(\bar{r}_i(t))\, e^{-i\omega_o t}\, e^{i(\bar{k}_s - \bar{k}_1)\cdot \bar{r}_i(t)}, \tag{8.1.2}$$

where I_o is the intensity of the light scattered from a particle placed at origin at the photodetector, and $P(\bar{r})$ is a weight dunction accounting for the size of the measuring volume, so the size of the measuring volume Ω_m is given by:

$$\Omega_m = \int |P(r)|^2 \, d^3r . \tag{8.1.3}$$

The number of particles at the time t, N(t), is obviously given by the sum:

$$N(t) = \sum_i |P(\bar{r}_i(t)|^2 , \tag{8.1.4}$$

and the mean number

$$\langle N(t) \rangle_P = \sum_i \langle |P(\bar{r}_i(t)|^2 \rangle_P . \tag{8.1.5}$$

The particle-particle correlation function is

$$\begin{aligned} \langle N(t)\ N(t+\tau) \rangle_P &= \Big\langle \sum_{i,j} |P(\bar{r}_i(t)|^2\ |P(r_j(t+\tau)|^2 \Big\rangle_P \\ &= \sum_i \langle |P(\bar{r}_i(t)|^2\ |P(\bar{r}_i(t+\tau)|^2 \rangle_P \\ &\quad + \langle N(t) \rangle_P \langle N(t+\tau) \rangle_P , \end{aligned} \tag{8.1.6}$$

where the last equation is obtained by using the assumption about the independence of the scattering particles.

For the second-order correlation function we then get:

$$g^{(2)}(t,\tau) = \frac{\langle E_s^*(t)\ E_s(t+\tau)\rangle_P}{(\langle E_s^*(t)\ E_s(t)\rangle_P\ \langle E_s^*(t+\tau)\ E_s(t+\tau)\rangle_P)^{\frac{1}{2}}}$$

$$= \frac{\langle \sum_{i,j} E_s^*(\bar{r}_i(t)\ E_s(\bar{r}_j(t+\tau)\rangle_P}{(\langle E_s^*(t)\ E_s(t)\rangle_P\ \langle E_s^*(t+\tau)\ E_s(t+\tau)\rangle_P)^{\frac{1}{2}}} \qquad (8.1.7)$$

$$= \frac{\langle \sum_{i} E_s^*(r_i(t)\ E_s(\bar{r}_i(t+\tau)\rangle_P}{(\langle E_s^*(t)\ E_s(t)\rangle_P\ \langle E_s^*(t+\tau)\ E_s(t+\tau)\rangle_P)^{\frac{1}{2}}},$$

where the last equation is obtained by using the fact that $\langle E_s(\bar{r}_i(t)\rangle = 0$, and the assumption of independence of the scattering particle. By inserting the expression for $E_s(\bar{r}_i(t))$ we get

$$g^{(2)}(t,\tau) = \frac{\langle \sum_i P\ (\bar{r}_i(t)P(\bar{r}_i(t+\tau))e^{+i(\bar{k}_i-\bar{k}_s)\cdot(\bar{r}_i(t)-\bar{r}_i(t+\tau))}\rangle P e^{-i\omega_o\tau}}{(\langle N(t)\rangle\ \langle N(t+\tau)\rangle)^{1/2}} \qquad (8.1.8)$$

For the fourth-order correlation we get:

$$g^{(4)}(t,\tau) = \frac{\langle E_s^*(t)\ E_s(t)\ E_s^*(t+\tau)E_s(t+\tau)\rangle_P}{\langle E_s^*(t)E_s(t)\rangle_P\ \langle E_s^*(t+\tau)E_s(t+\tau)\rangle_P} \qquad (8.1.9)$$

$$= \frac{\langle \sum_{i,j,k,l} E_s^*(\bar{r}_i(t))E_s(\bar{r}_j(t))E_s(r_k(t+\tau))E_s(\bar{r}_l(t+\tau))\rangle_P}{\langle E_s^*(t)\ E_s(t)\rangle_P\ \langle E_s^*(t+\tau)\ E_s(t+\tau)\rangle_P}$$

In the four-fold sum only terms where

$i = j = k = l$

and

$l = j \neq k = l$

and

$i = l \neq j = k$

will contribute to the sum, and by inserting the expression for $E_s(\bar{r}_i(t))$ we find:

$$g^{(4)}(t,\tau) = \frac{\left\langle \sum_i |P(\bar{r}_i(t))|^2 \; |P(\bar{r}_i(t+\tau))|^2 \right\rangle_P}{\langle N(t)\rangle_P \langle N(t+\tau)\rangle_P}$$

$$+ \frac{\left\langle \sum_{i\neq l} |P(\bar{r}_i(t))|^2 |P(r_l(t+\tau))|^2 \right\rangle_P}{\langle N(t)\rangle_P \langle N(t+\tau)\rangle_P} \qquad (8.1.10)$$

$$+ \left\langle \sum_{i\neq k} \frac{P^*(\bar{r}_i(t)) P(\bar{r}_i(t+\tau)) P^*(\bar{r}_k(t+\;)) P(\bar{r}_k(t))}{\langle N(t)\rangle_P \langle N(t+\tau)\rangle_P} \right.$$

$$\times \left. e^{i(\bar{k}_i - \bar{k}_s)(\bar{r}_i(t) - r_i(t+\tau) + \bar{r}_k(t+\tau) - \bar{r}_k(t))} \right\rangle_P$$

and further by insertion of the particle-particle correlation function:

$$g^{(4)}(t,\tau) = \frac{\langle N(t)\; N(t+\tau)\rangle_P - \langle N(t)\rangle_P \langle N(t+\tau)\rangle_P}{\langle N(t)\rangle_P \langle N(t+\tau)\rangle_P} \qquad (8.1.11)$$

$$+ 1 + |g^{(2)}(t,\tau)|^2 .$$

We see that the finite extent of the measuring volume gives rise to an additional term in the Siegert relation, which represents the particle number fluctuation in the measuring volume. If we can assume that the number of particles has a Poisson distribution, this term varies as $1/\langle N\rangle$ for "small" values of the time displacement τ. This means that the Siegert relation holds for, say, $\langle N\rangle \gtrsim 10$, a relation which we could expect from the central limit theorem.

8.2. Brownian Motion

As a first example, we consider particles which undergo Brownian motion in a fluid. The Brownian motion is due to the collision between the macroscopic particles and the molecules in the surrounding medium. To calculate the second- and fourth-order correlation functions, we must know the joint probability density for a particle at a time t and for the same particle at a time τ later; we write it as:

$$S(\bar{r}_2(t),\; \bar{r}_1(t+\tau)) ,$$

and we can express this quantity as

$$S(\bar{r}_1(t),\ \bar{r}_1(t+\tau)) = S(\bar{r}_1(t))\ S(\bar{r}_1(t+\tau)\ |\ r_1(t)), \tag{8.2.1}$$

where $S(r_1(t))$ is the probability density for a particle at a time t, and $S(r_1(t+\tau)\ |\ r_1(t))$ is a conditional probability density; i.e. the probability that the particle is at a position $r_1(t+\tau)$ under the condition that it was at the position $r_1(t)$ at the time t. Now, by assuming stationarity and that the particles are uniformly distributed in the space, we can write:

$$S(r_1(t)) = \frac{1}{\Omega_n} \tag{8.2.2}$$

and

$$S(r_1(t+\tau)\ |\ r_1(t)) = F(r_1(t+\tau) - r_1(t), \tau), \tag{8.2.3}$$

where Ω_n is the volume in which the particles are distributed. The function $F(\Delta r, \tau)$ is a Gaussian distribution[36], a fact which is obvious if we think of a large collection of particles and ask for the probability that a particle is displaced a distance Δr from its initial position in the time τ. We obtain the answer and use the central limit theorem:

$$F(\Delta r, \tau) = (2/3\pi \langle (\Delta r)^2 \rangle)^{-3/2}\ e^{-\frac{3(\Delta r)^2}{2\langle (\Delta r)^2 \rangle}} \tag{8.2.4}$$

where $\langle (\Delta r)^2 \rangle$ is the variance of the displacement Δr. To determine the variance of the displacement, we must consider the equation of motion for the particles. The forces which act on the particles are separated into two parts, an average force represented by the friction of the surrounding fluid and a fluctuating force due to the pressure fluctuation in the fluid, and we obtain the Langevin equation:

$$m\ddot{r} = -m\beta\dot{r} + mF(t), \tag{8.2.5}$$

where r is the particle position and m its mass, F(t) is the fluctuating force, and β is a reciprocal relaxation time. For spherical particles of radius a, β is generally given by the Stoke's equation:

$$m\beta = 6\pi a \eta, \tag{8.2.6}$$

where η is the viscosity of the fluid. In this text the Stoke's equation is used, but it should be pointed out that a more exact form of the Stoke's equation yields[37]:

$$m\beta = 6\pi a\eta \left(1 + \frac{3}{8}\rho\frac{av}{\eta}\right),$$

where ρ is the density of the fluid and v the particle velocity. The above equation is valid under the condition that $2av\rho/\eta \ll 1$, and further that the particle size must be great in relation to the coherence length of the molecules of the fluid.

$$\langle(\Delta r)^2\rangle = \left\langle \int_0^t \int_0^t v(t')\ v(t'')\ dt'\ dt'' \right\rangle. \qquad (8.2.7)$$

By differentiating $\langle(\Delta t)^2\rangle$ twice and using the Langevin equation, we obtain the equation:

$$\ddot{S} = 2\langle v(t)^2\rangle - \beta\dot{S} + \int_0^t \langle v(t')\ F(t)\rangle\ dt', \qquad (8.2.8)$$

where $S = \langle(\Delta r)^2\rangle$. The last term on the right will vanish because both $\langle v(t')\rangle$ and $\langle F(t)\rangle$ are zero and uncorrelated. The mean square velocity is equal to

$$\langle v(t)\rangle^2 = \frac{3k_BT}{m},$$

and we obtain:

$$\ddot{S} = \frac{6k_BT}{m} - \beta\dot{S}.$$

By solving this equation we finally get:

$$\langle(\Delta r)^2\rangle = \frac{6k_BT}{\beta^2 m}\left(\beta t - 1 + e^{-\beta t}\right) \qquad (8.2.9)$$

a result obtained by Chandrasekhar[38] (1943).

Now, returning to our aim to calculate the correlation function for the scattered field, we can with the above equations in mind, write the second order correlation function as:

$$g^{(2)}(\tau) = e^{-i\omega_o\tau} \frac{\sum_i \int P^*(\bar{r}_1)P(\bar{r}_2)e^{i\bar{\Delta k}\cdot(\bar{r}_1-\bar{r}_2)} S(\bar{r}_1)F(\bar{r}_2-\bar{r}_1,\tau)d^3\bar{r}_1 d^3\bar{r}_2}{\sum_i \int |P(\bar{r}_1)^2|\, S(\bar{r}_1)d^3\bar{r}_1}$$

$$= e^{-i\omega_o\tau} \frac{1}{\Omega_m} \int P^*(\bar{r}_1)\, P(\bar{r}_2)\, e^{i\Delta k\cdot(\bar{r}_1-\bar{r}_2)}\, F(\bar{r}_2-\bar{r}_1,\tau)d^3\bar{r}_1 d^3\bar{r}_2$$

where $\Delta\bar{k} = \bar{k}_i - \bar{k}_s$.

Now, if we assume for simplicity that the measuring volume can be described by a Gaussian weight function, i. e.

$$P(r) = e^{-r^2/4\sigma_o^2}, \tag{8.2.11}$$

where σ_o is determined by the relation

$$\Omega_m = (2\pi\sigma_o^2)^{3/2},$$

we then get by insertion of $P(\bar{r})$:

$$g^{(2)}(\tau) = \frac{1}{\left(1 + \frac{1}{12}\frac{\langle(\Delta r)^2\rangle}{\sigma_o^2}\right)^{3/2}}$$

$$\times \exp\left\{-2(\sigma_o\Delta k)^2 \frac{\frac{1}{12}\frac{\langle(\Delta r)^2\rangle}{\sigma_o^2}}{1 + \frac{1}{12}\frac{\langle(\Delta r)^2\rangle}{\sigma_o^2}} - i\omega_o\tau\right\}. \tag{8.2.12}$$

From the expression for $g^{(2)}(\tau)$ we see that if the quantity $\sigma_o\Delta k$ fulfils the condition:

$$\sigma_o\Delta k \gg 1, \tag{8.2.13}$$

then $g^{(2)}(\tau)$ is only non-vanishing for τ values that fulfil the inequality

$$\langle(\Delta r)^2\rangle \ll 12\sigma_o^2. \tag{8.2.14}$$

In this case, $g^{(2)}(\tau)$ is practically independent of the measuring volume and we can write

$$g^{(2)}(\tau) = e^{-\frac{1}{6}(\Delta k)^2 \langle(\Delta r)^2\rangle} e^{-i\omega_o \tau}. \tag{8.2.15}$$

The quantity $\sigma_o \Delta k$ can be interpreted as the number of virtual planes in the measuring volume and it will in general be greater than one in experimental set-ups.

The coherence time τ_c (to second order) of the scattered light can be found by setting:

$$\frac{1}{6}(\Delta k)^2 \langle(\Delta r)^2\rangle = 1 \tag{8.2.16}$$

or

$$\frac{k_B T}{m\beta^2}(\Delta k)^2 (\beta\tau_c - 1 + e^{-\beta\tau_c}) = 1 .$$

The term $e^{-\beta\tau}-1$ can be neglected; the quantity γ

$$\gamma = \frac{k_B T}{m\beta^2}(\Delta k)^2$$

will in practice fulfil the unequality $\gamma \ll 1$, and we get:

$$\tau_c = \frac{m\beta}{k_B T(\Delta k)^2} = \frac{1}{D(\Delta k)^2}, \tag{8.2.17}$$

where D is the diffusion constant

$$D = \frac{k_B T}{m\beta} .$$

For the second-order correlation function we then get

$$g^{(2)}(\sigma) = e^{-D(\Delta k)^2 |\tau|} . \tag{8.2.18}$$

By Fourier-transforming the second-order correlation function, we obtain the spectral density of the scattered field. In general, the spectral density is related to the incomplete gamma function, but for the case where

$\gamma \ll 1$ it can be approximated by a Lorentzian in the frequency range

$$|\omega| < \left(\frac{k_B T}{m}\right)^{1/2} \Delta k \,.$$

The variance of the spectral density can be calculated in general from eq. (8.4)

$$\langle(\Delta\omega)^2\rangle = \frac{k_B T}{m} (\Delta k)^2 \left(1 + \frac{3}{4} \frac{1}{(\sigma_o \Delta k)^2}\right) \tag{8.2.19}$$

$$= \langle(\Delta k \cdot v)^2\rangle \left(1 + \frac{3}{4} \frac{1}{(\sigma_o \Delta k)^2}\right) ,$$

which is independent of β.

The last result is maybe of academic interest only, but it illustrates that however close the spectral density comes to the pure Lorentzian, where the second-order moment does not exist, the spectral density will still have a finite second-order moment.

The contribution to the fourth-order correlation function from the particle fluctuation in the measuring volume is

$$\frac{\langle \Delta N(t)\, \Delta N(t+\tau)\rangle}{\langle N(t)\rangle^2} = \frac{1}{\langle N(t)\rangle^2} \sum_i \langle |P(\bar{r}_1(t)|^2 \, |P(\bar{r}_2(t+\tau)|^2\rangle$$

$$= \frac{1}{n_o \Omega_m} \frac{1}{\Omega_m} \int |P(\bar{r}_1)|^2 \, |P(\bar{r}_2)|^2 F(\bar{r}_2 - \bar{r}_1, \tau)\, d^3\bar{r}_1 d^3\bar{r}_2 \tag{8.2.20}$$

$$= \frac{2^{-3/2}}{n_o \Omega_m} \left(1 + \frac{1}{6} \frac{\langle(\Delta r)^2\rangle}{\sigma_o^2}\right)^{-3/2}$$

where n_o is the particle concentration. The factor $2^{-3/2}$ results from the assumption of a Gaussian measuring volume. For the fourth-order correlation function we then get:

$$g^{(4)}(\tau) = \frac{2^{-3/2}}{n_o \Omega_m} \left(1 + \frac{1}{6} \frac{\langle(\Delta r)^2\rangle}{\sigma_o^2}\right)^{-3/2} + 1 + |g^{(2)}(\tau)|^2 \tag{8.2.21}$$

The decay constant of the particle fluctuation is found by setting

$$\frac{1}{6} \frac{\langle(\Delta r)^2\rangle}{\sigma_o^2} = 1 ,$$

and we get

$$\tau_{cp} = \frac{m\beta\sigma^2}{k_B T} = \tau_c (\sigma_o \Delta k)^2 , \tag{8.2.22}$$

If there is a great number of virtual interference lines, we see that the decay of the particle number fluctuations is much slower than the decay of the second-order correlation function. The effect of the number fluctuation has been experimentally investigated by Schaefer et al.[39].

To summarize, we find that information can be obtained about the translational diffusion constant for spherical macroscopic particles by measuring the second- and fourth-order correlation functions. Experiments of this kind were first performed by Cummins et al.[40] for ordinary particles and the method is at present widely used to obtain information about the diffusion properties of all kinds of particles (i.e. biological molecules[41] and self-propelled organisms[42].

8.3. Light Scattering from Particles Suspended in Laminar and Turbulent Flows

Velocity measurement of moving fluids is to-day a well-established technique, known as Laser-Doppler anemometry. By measuring the light scattered from macroscopic particles suspended in a moving fluid, one can obtain information about the velocity of the fluid, the degree of turbulence and the spatial structure of the velocity field. A basic assumption in these measurements and the interpretation of their results is that the particles are exactly followed by the motion of the fluid; that is, if the velocity of the fluid is denoted $U(r, t)$ and the velocity of the particle number k is denoted $v_k(t)$, then:

$$v_k(t) = U(r_k(t), t) .$$

The assumption is generally said to be justified when the size of the particles is small compared with the smallest scale of the spatial variation of the fluid velocity[8,10], but we wish to point out that the consequences of this condition do not necessarily lead to the conclusion that the particles are exactly followed by the fluid elements. Actually, the motion of the particles will be a movement with the fluid elements superimposed with a Brownian motion of the particles, with the above condition fulfilled. To take the effect of Brownian motion into account, we use the same method as in part 8.2, in which we find a Langevin equation for the relative motion of the particles, and as before assume that the initial position of the particles is independent and that we are dealing with stationary processes.

By denoting the relative and absolute velocity of the particles $v^{(r)}(t)$ and $v^{(a)}(t)$ respectively, we have

$$v^{(r)}(t) = v^{(a)}(t) - U(r, t) , \qquad (8.3.1)$$

and the Langevin equation becomes

$$\dot{v}(r) = -\beta \dot{v}(r) + F(t) , \tag{8.3.2}$$

where F(t) is a fluctuation force, due to the pressure fluctuation in the fluid.

From 8.1 we have for the second-order correlation function

$$g^{(2)}(\tau) = e^{-i\omega_o \tau} \frac{1}{\zeta_m} \int P^*(\bar{r}_1) P(\bar{r}_2) e^{i\bar{k}\cdot(\bar{r}_1 - \bar{r}_2)} S(\bar{r}_2, \bar{r}_1, \tau) d^3\bar{r}_1 d^3\bar{r}_2, \tag{8.3.3}$$

and we wish to find an expression for the conditional probability density $S(\bar{r}_1 \mid \bar{r}_2, \tau)$ in the case of a moving fluid. As in the Brownian case, the relative displacement will have a Gaussian distribution, and by assuming isotropicality in the medium we have

$$S(\bar{r}_2 \mid \bar{r}_1 \tau) = (2/3\pi\sigma^2)^{-3/2} \exp\left(\frac{-3(\bar{r}_2 - \bar{r}_1 - \langle \Delta\bar{r} \rangle_M)^2}{2\sigma^2} \right) \tag{8.3.4}$$

where

$$\sigma^2 = V(\Delta r)$$

and

$$\langle \Delta r \rangle_M = \left\langle \int_o^\tau \bar{v}^a(t) dt \right\rangle_M . \tag{8.3.5}$$

The averaging is an ensemble-averaging over the statistical properties of the medium, and the result will in general depend on the initial position r_1. For the mean of absolute velocity, we obtain from the Langevin equation:

$$\langle v^a(t) \rangle_M = \langle U(r(t), t) \rangle_M = \langle U(r(o), o) \rangle \tag{8.3.6}$$

and for the displacement r

$$\langle \Delta\bar{r} \rangle_M = \langle U(\bar{r}_1, o) \rangle \tau .$$

The variance of Δr becomes

$$\sigma^2(\bar{r},\tau) = \mathbf{V}(\Delta r) = \left\langle \left(\int_0^\tau (v^a(t) - \langle v^a \rangle) dt \right)^2 \right\rangle$$

$$= \int_0^\tau \int_0^\tau \langle v^{(r)}(t_1) v^{(r)}(t_2) \rangle dt_1 dt_2$$

$$+ \int_0^\tau \int_0^\tau \langle (U(\bar{r}_1(t_1),t_1) - \langle U(\bar{r}_1(t_1),t_1)\rangle_M)(U(r_1(t_2),t_2)$$

$$- \langle U(r_1(t_2),t_2)\rangle_M)\rangle_M dt_1 dt_2 \qquad (8.3.7)$$

$$= \langle v^{(r)}(o)^2 \rangle_M \quad (\beta\tau - 1 + e^{-\beta\tau})/\beta^2$$

$$+ \langle (\Delta U)^2 \rangle_M \int_0^\tau \int_0^\tau R_L(t_2 - t_1) dt_1 dt_2 \,,$$

where we have introduced

$$\langle (\Delta U)^2 \rangle_M = \langle U(\bar{r}_1(t_1),t_1) - \langle U(\bar{r}_1(t_1),t_1)\rangle_M)^2 \rangle_M \qquad (8.3.8)$$

and the Lagrangian correlation function $R_L(t_2 - t_1)$

$$R_L(t_2 - t_1) = \frac{\langle \Delta U(\bar{r}_1(t_1),t_1) \Delta U(\bar{r}_1(t_2),t_2)\rangle_M}{\langle (\Delta U)^2 \rangle_M} \,. \qquad (8.3.9)$$

$\langle (\Delta U)^2 \rangle$ and $R_L(t_2 - t_1)$ will in general depend on the initial position r_1. In the above calculation we have assumed that the pressure fluctuations are uncorrelated with the velocity fluctuation in the fluid.

Ordinarily, the moving fluid is contained in pipes, and if we measure close to the walls, the walls will influence the correlation function. For simplification, we will assume that our measuring volume is so far from the walls that this effect is negligible. Further, we will assume that the velocity gradient in the measuring volume is small, so we neglect the dependence on the initial position for the quantities $\langle U(\bar{r}_1,\tau)\rangle$ and $\sigma^2(\bar{r},\tau)$ by setting:

$$\langle U(\bar{r},t)\rangle = \langle U(\bar{r}_1,o)\rangle$$

and

$$v(\bar{r},\tau) = \sigma(\bar{r}_1,\tau),$$

where $\bar{r}_1$ is a mean point in the measuring volume.

Now, with this assumption in mind, we obtain after some calculations:

$$g^{(2)}(\tau) = \frac{1}{(1+\delta^2)^{3/2}} \cdot e^{-\frac{\sigma^2(\Delta k)^2}{6(1+\delta^2)} - \frac{\langle U^2\rangle \tau^2}{8\sigma_o^2(1+\delta^2)} + \frac{i\Delta k\langle U\rangle \tau}{1+\delta^2} - i\omega_o\tau} \tag{8.3.10}$$

where

$$\delta^2 = \frac{1}{12}\,\frac{\sigma^2(\bar{r}_1,\tau)}{v_o^2},$$

and again, if the number of virtual interference planes is much greater than a unit, which we will assume, we can neglect δ^2, and finally we obtain for the second-order correlation function:

$$g^{(2)}(\tau) = e^{-\frac{\sigma^2(\bar{r},\tau)(\Delta k)^2}{6} - \frac{\langle U\rangle^2\tau^2}{8\sigma_o^2} - i(\omega_o - \Delta k\langle U\rangle)\tau}. \tag{8.3.11}$$

To interpret this expression, we consider the terms in the exponential at eq. 8.3.11. The term

$$\frac{\langle U^2\rangle \tau^2}{8\sigma_o^2} \tag{8.3.12}$$

is a decay term due to the finite transit time of the particle in the measuring volume. If we define the transit time τ_o as

$$\tau_o = \frac{2\sigma_o}{\langle U\rangle}, \tag{8.3.13}$$

we can write eq. 8.3.12 as:

$$\frac{\langle U^2\rangle \tau^2}{2\sigma_o^2} = \frac{\tau^2}{2\tau_o^2}$$

and this gives rise to a broadening of the spectrum of $g^{(2)}(\tau)$ that is termed the Doppler radar ambiguity broadening[8,11]. The decay term

$$1/6\ \sigma^2(\bar{r},\sigma)(\Delta k)^2 = 1/2\,\Delta\bar{k}\cdot\langle(\bar{r}-\langle\Delta r\rangle)(\Delta r-\langle\Delta r\rangle)\rangle\cdot\Delta\bar{k} \qquad (8.3.14)$$

is due to the fluctuation of the particle position. The decay time τ_1 of this term can be defined by the relation

$$1/3\ \sigma^2(\bar{r},\tau_1)(\Delta k)^2 = 1 \qquad (8.3.15)$$

τ_1 reflects how fast the particle position is spread out in the $\Delta\bar{k}$ direction within a length of

$$1/\Delta k = \lambda/2\pi ,$$

where λ is the distance between the virtual interference planes. Further from eq. (8.3.7), we see that $\sigma^2(r,\tau)$ is composed of two parts, one due to the diffusional movement and one due to the turbulent motion. The time scale β in the diffusional motion is of the order of

$$\beta^{-1} \approx 10^{-9}\ \mathrm{s}$$

and the diffusion constant D is of the order of

$$D = \frac{k_B T}{m\beta} \approx 10^{-8}\ \mathrm{cm}^2/\mathrm{s}$$

so, for ordinary fluid velocities, the short-time behaviour ($\beta\,\tau \ll 1$) gives no contribution to the decay of the correlation function $g^{(2)}(\tau)$. The long-time behaviour of $\sigma^2(r,\tau)$, due to the diffusional motion, varies as

$$1/6\ \sigma^2(r,\tau) = (\Delta k)^2 D\tau ,$$

and the decay constant becomes

$$\tau_{10} = 1/D\,(\Delta k)^2 \approx 0.1\ \mathrm{s}.$$

In the absence of turbulent motion, we then find for $g^{(2)}(\tau)$

$$g^{(2)}(\tau) = e^{-D(\Delta k)^2|\tau|}\ e^{-\frac{\langle U\rangle^2}{8\sigma_o^2}\tau^2}\ e^{-i(\omega_o-\Delta k\langle\bar{U}\rangle)\tau} . \qquad (8.3.16)$$

The spectrum of $g^{(2)}(\tau)$ becomes a Gaussian when the velocity fulfils the condition

$$\langle U \rangle \; \rangle\rangle \; 2\sigma_o(\Delta k)^2 D \; , \tag{8.3.17}$$

and the information about the diffusional properties of the medium is in practice lost, despite the fact that the variance of the spectrum calculated from eq. (8.3.10) is given by:

$$\langle (\Delta\omega)^2 \rangle = - \frac{d^2}{d\tau^2} \mid g^{(2)}(\tau) \mid_{\tau=0} = 1/3(\Delta k)^2 \langle (v^{(r)})^2 \rangle + \frac{\langle U^2 \rangle}{4\sigma_o^2}$$

$$= D(\Delta k)^2 \beta \left(1 + \frac{\langle U \rangle^2}{4\beta D(\Delta k \sigma_o)^2}\right) \tag{8.3.18}$$

and though the condition stated in eq. 8.3.17, for $\langle U \rangle$ is fulfilled, the variance will be independent of $\langle U \rangle$ if

$$\langle U \rangle \; \langle \; (4\beta D(\Delta k_{\;o})^2)^{1/2} \; .$$

This could be considered a paradox, but there is nothing to prevent the energy in a spectrum concentrating into a small area, though the spread of the spectrum is much greater than this area or goes to infinity as in the case of a Lorentzian spectrum.

Now, when the mean value of the velocity $\langle U \rangle$, fulfils the condition

$$\langle U \rangle \lesssim 2\sigma_o(\Delta k)^2 D \tag{8.3.19}$$

the spectrum is non-Gaussian, a case first shown by Edwards et al, [43]. The spectrum will in this case be related to the error function.

In an experimental situation where we measure on a laminar flow at low velocities and obtain a spectrum of the scattered light of the form just described, we must use a curve-fitting procedure to decide which of the two cases described we are dealing with. If we try to measure the variance of the obtained spectra, and compare it with the calculated variance in eq. (8.3.18), only information on the bandwidth of the spectrum analyser will be obtained.

In the case of turbulent motion, we can neglect the diffusional motion and we have from eq. (8.3.7):

$$\sigma^2(\bar{r},\tau) = \langle (\Delta U)^2 \rangle \int_0^\tau \int_0^\tau R_L(t_2-t_1)\, dt_2 dt_1$$

$$= 2\langle (\Delta U)^2 \rangle \, \tau \int_0^\tau (1-t/\tau) R_L(t)\, dt. \qquad (8.3.20)$$

If we denote τ_L the correlation time for the Lagrangian correlation function $R_L(\tau)$, we can distinguish between the two cases, one where τ is smaller than τ_L and one where τ is greater than τ_L. In the case of short-time behaviour of $\sigma^2(r,\tau)$, where

$$\tau \ll \tau_L,$$

we have

$$\sigma^2(r,\tau) \approx 2\langle (\Delta U)^2 \rangle R_L(\sigma) \int_0^\tau (\tau - t)\, dt = \langle (\Delta U)^2 \rangle \tau^2, \qquad (8.3.21)$$

which is the same τ dependence as in the short-time behaviour of the diffusional motion.

For the long-time behaviour of $\sigma^2(r,\tau)$

$$\tau \gg \tau_L$$

we get

$$\sigma^2(r,\tau) = 2\langle (\Delta U)^2 \rangle \tau \int_0^\infty (1-t/\tau) R(t)\, dt$$

$$\approx 2\langle (\Delta U)^2 \rangle \left(\int_0^\infty R(t)\, dt \right) \tau. \qquad (8.3.22)$$

The time scale of τ_L can be estimated by introducing a length, l_L, which represents a typical length in the system, for example, the shear dimension of the flow system. τ_L and l_L is related through the relation

$$l_L = \langle (\Delta U)^2 \rangle^{1/2} \tau_L. \qquad (8.3.23)$$

In the case of short-time behaviour, we obtain the decay time τ_1 by using eq. (8.3.20)

$$\tau_1 = \left(\frac{3}{\langle (\Delta U)^2 \rangle (\Delta k)^2} \right)^{1/2} = \frac{\sqrt{3}\, \tau_L}{(\Delta k\, l_L)}. \qquad (8.3.24)$$

and to satisfy the condition for short-time behaviour, l_L must fulfil the condition:

$$\Delta k \, l_L \gg 1 .$$

l_L will usually be of the order of 1 cm, so the above condition will ordinarily be fulfilled, except in cases where the scattering angle (see fig. 8.1) is small.

The second-order correlation function will in this case be Gaussian

$$g^{(2)}(\tau) = e^{-\frac{1}{6}(\Delta k)^2 \langle (\Delta U)^2 \rangle \tau^2 - \frac{\langle U \rangle^2 \tau^2}{8\sigma_o^2}} e^{i(\omega_o - \Delta k \langle \bar{u} \rangle)\tau} \qquad (8.3.25)$$

and this spectral density of the light is a Gaussian function with a variance of

$$\langle (\Delta\omega) \rangle^2 = \Delta k \cdot \langle \Delta \bar{U} \Delta \bar{U} \rangle \cdot \Delta k + \frac{\langle U \rangle^2}{4\sigma_o^2} . \qquad (8.3.26)$$

To summarize, the spectrum of $g^{(2)}(\tau)$ contains information about the mean velocity and the turbulence intensity in the measuring volume, with the restriction stated above and the turbulence and diffusional properties of a fluid can be described in a similar way. With respect to the fourth-order correlation function in the case of turbulence, we will not go into detail but refer to a paper by Bertolotti et al.[44], where it is shown that the Siegert relation is not fulfilled in general and that the fourth-order correlation function contains information about the spatial variation of the velocity fluctuation.

9. BRILLOUIN SCATTERING

9.1. Theoretical Investigation of Light Scattering from Atoms and Molecules

An incident light beam in a medium will give rise to electric polarisation. A dipole-moment will be induced on the single molecules in the medium and, together with the remaining molecules, this moment gives the electric polarisation. If the particles in the medium were at fixed positions, in the same thermodynamical environments, and furthermore all of the same kind, it is obvious that the electric polarisation would be coherent with the incident light beam, that the total electric field would have the same direction of propagation and the same polarisation as the incident light beam,

and that no light scattering would occur. Another way to explain this fact is that it is always possible to find two particles placed so that the scattered field from them is out of phase, which implies that the field would vanish. The effect of light scattering is therefore a result of the inhomogeneities in the optical properties of the medium, as, for instance, fluctuations in the density of the particles.

In this part we will investigate the effect of density fluctuation on the scattered light field in an optical isotropic medium composed of molecules or atoms all of the same kind. We deal with two cases, one where the scattering particles can be considered to move as a kinetic gas, and one where the movement and interaction of the particles can be described in a hydrodynamical approach. In the latter case, the effect of Brillouin scattering will appear. An experimental investigation of Brillouin scattering has been made, and the equipment used and the results obtained are described at the end of this chapter.

A basic formula in this part is the expression for the scattered light from a collection of particles. From eq. B.12

$$E_{ks}(\bar{r},\omega) = -\frac{1}{8\pi^2\varepsilon_o}\frac{e^{ikr}}{r}\,k_s \times (\bar{k}_s \times \qquad (9.1.1)$$

$$(\int \alpha(\omega')\,\bar{E}_o(r',\omega')\,\Delta n(r',\omega-\omega')\,e^{-i\bar{k}_s\cdot r'}\,d\omega' dr'$$

where E_o is the incident light field, $\alpha(\omega)$ is the polarisability of the molecules, Δn is the fluctuation part of the number density of the molecules, and $\bar{k}_s$ is given by

$$\bar{k}_s = k\,\bar{r}/r\,,$$

where $\bar{r}$ is the point of observation.

By assuming that the incident light field is a plane monochromatic wave, with a frequency ω_o, we can write the scattered field as:

$$\bar{E}_{k_s}(\bar{r},t) = -\frac{\alpha(\omega_o)}{4\pi\varepsilon_o}(\hat{k}_s \times(\hat{k}_s \times \hat{\varepsilon}\,))\frac{e^{ikr}}{r}e^{-i\omega_o t}\int \Delta n(r',t) \qquad (9.1.2)$$

$$\times\, e^{i(\bar{k}_i-\bar{k}_s)\cdot r'}\,d^3r'$$

where $\hat{\varepsilon}$ and $\bar{k}_i$ are the amplitude and wavevector, respectively, of the incident light beam.

From eq. 9.1.2 it is seen that the scattered field is proportional to the Fourier transformed of the fluctuating part of the number density in

the direction $\Delta\bar{k} = \bar{k}_i - \bar{k}_s$, which means that only fluctuations in this direction will contribute to the scattered field.

By denoting the Fourier transformed of the number density ρ_k, i.e.,

$$\rho_{\bar{k}}(t) = \int n(\bar{r},t)\ e^{-i\bar{k}\cdot\bar{r}}\ dr^3, \tag{9.1.3}$$

we obtain for the second-order correlation function of the scattered field:

$$\begin{aligned} R_{k_s}(\tau) &= \langle E_{\bar{k}_s}(t)\ E_{\bar{k}_s}(t+\tau)\rangle \\ &= I_o\left|\frac{k^2\alpha(\omega_o)\sin\phi}{4\pi\varepsilon_o r}\right|^2 \langle\rho_{\Delta\bar{k}}(t)\ \rho^*_{\Delta\bar{k}}(t+\tau)\rangle \end{aligned} \tag{9.1.4}$$

where ϕ is the angle between the direction of observation and the direction of the incident electric field, and I_o is the intensity of the incident light beam.

To find an expression for the correlation function of the ρ_k's, we perceive the number density as a sum of delta functions, i.e.,

$$n(r,t) = \sum \delta(r-r_i(t)) \tag{9.1.5}$$

and, by insertion of this relation into eq. 9.1.3, we obtain

$$\rho_{\bar{k}}(t) = \sum_i e^{-i\bar{k}\cdot\bar{r}_i(t)} \tag{9.1.6}$$

and further

$$\begin{aligned} &\langle\rho_{\Delta\bar{k}}(t)\ \rho^*_{\Delta k}(t+\tau)\rangle \\ &= \Big\langle \sum_{i,j} e^{-i\Delta k\cdot(\bar{r}_i(t)-\bar{r}_j(t+\tau)} \Big\rangle \\ &= \Big\langle \sum_i e^{-i\Delta\bar{k}\cdot(\bar{r}_i(t)-r_i(t+\tau))} \Big\rangle + \Big\langle \sum_{i,j}{}' e^{-i\Delta\bar{k}\cdot(\bar{r}_i(t)-\bar{r}_j(t+\tau))} \Big\rangle . \end{aligned} \tag{9.1.7}$$

To interpret the two terms in eq. (9.1.7), we assume that the particles are equally distributed in a volume V and that we deal with stationary processes in time and space .

By introducing the self-conditional probability function $F_s(r_1 - r_o, \tau)$ - which denotes the probability of finding a particle at the position r_1 and at the time τ when the particle was initially at the position r_o - and also the

conditional probability $F_p(r_1 - r_0, \tau)$ of finding a particle at the position r_1 and at the time τ when a second particle was initially at the position r_0, we can obtain from eq. (9.1.7)

$$\langle \rho_{\Delta k}(t)\,(t)\,\rho^*_{\Delta k}(t+\tau)\rangle = \frac{N}{V} \int F_s(\bar{r},) \, e^{-i\Delta \bar{k}\cdot\bar{r}} \, d^3r$$

$$+ \frac{N(N-1)}{V} \int F_p(\bar{r},\tau) \, e^{-\Delta\bar{k}\cdot\bar{r}} \, d^3r \qquad (9.1.8)$$

where N is the number of particles in the volume V. The self-conditional probability function gives information about the way a single particle interacts with the remaining particles, whereas the conditional probability F_p gives information about how a pair of particles interacts with the remaining particles. The function $F_s(r,\tau)$ will be a delta function in space when $\tau = 0$, and continue to be a very sharply peaked function up to the time of free flight τ_f, where the function begins to smear out. This expresses the fact that the initial position and the position at the time τ are highly correlated until collisions with neighbouring particles begin to take place. The pair conditional probability $F_p(r,\tau)$ has a more complicated behaviour. It is clear that if the particles moved without interaction F_p would be constant and would give no contribution to eq. (9.7). If there is interaction between the particles there must exist a small region about the initial position for $\tau = 0$, where the probability to find another particle must be practically zero (the interaction is repulsive at short distances). This region must be determined by the diameters of the molecules. Far away from the initial position we must expect that F_p becomes constant and equal to $1/V$. In areas between the region defined above and this region, we must expect that F_p exceseds $1/V$ due to the attractive part of the interacting forces. For time displacements smaller than the time of free flight, the particle that was sited at the initial position will have a position that is distributed according to $F_s(r,\tau)$ and the picture stated above for $\tau = 0$ will be smeared out. With the onset of the effect of collisions, the picture will change and particles far from the initial position will be affected and the pair probability at these points will differ from $1/V$. For time displacement much greater than the time of free flight the position of these points will be given by

$$r = c\tau$$

where c is the velocity for which a message can be transmitted in the medium, i.e. the velocity of sound in the medium. In figs. 9.1 and 9.2,

F_s and F_p are sketched for different values of τ. τ_f denotes the time of free flight and l_f the mean free paths. A more quantitative discussion of the behaviour of F_s and F_p can be found in [45].

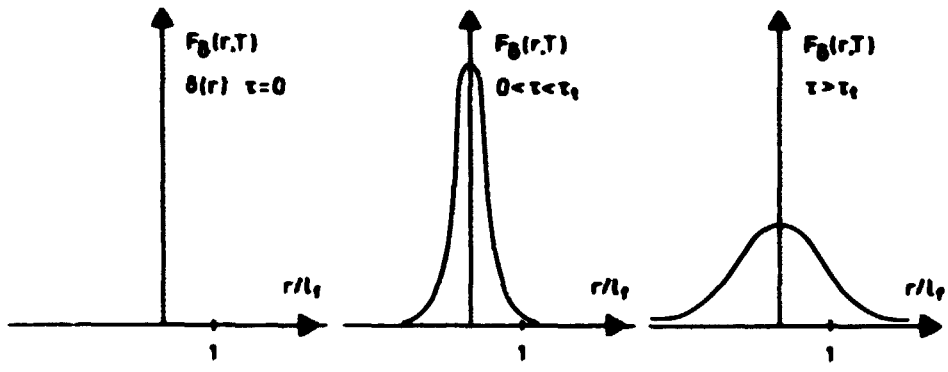


Fig. 9.1

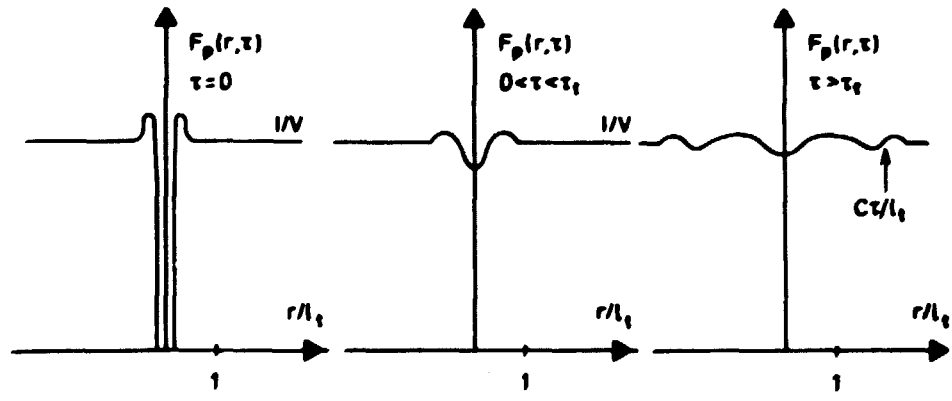


Fig. 9.2

In eq. 9.1.8 we deal with the Fourier transformed of F_s and F_p, and by means of $\Delta\bar{k} = \bar{k}_i - \bar{k}_s$ and the mean free path l_f, we can distinguish between two characteristic cases that are defined in the following.

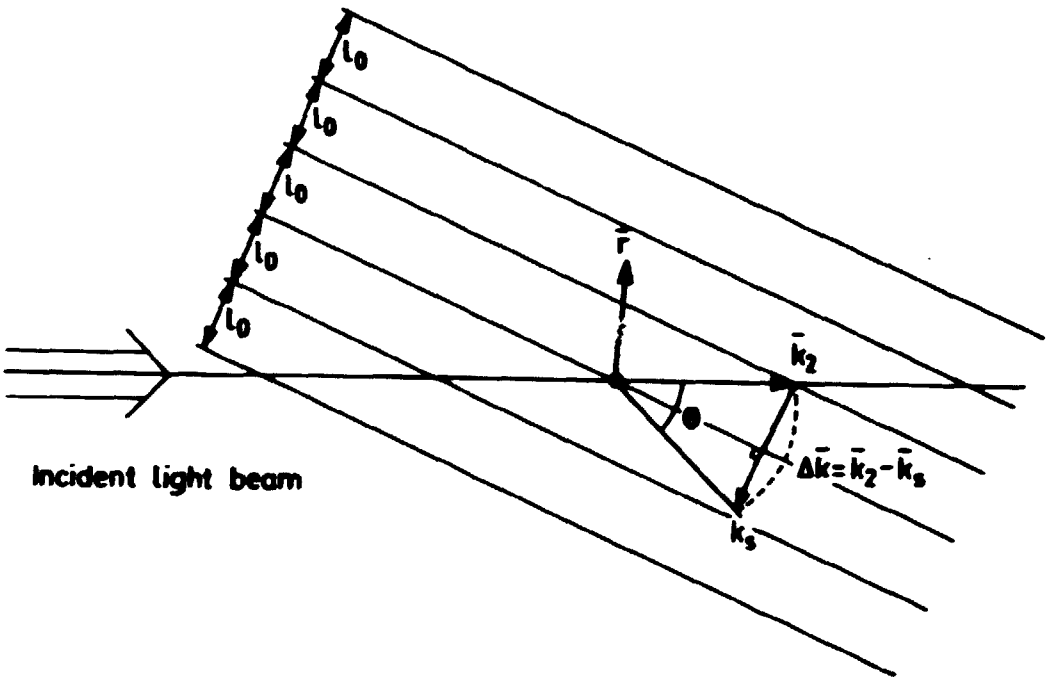


Fig. 9.3

Figure 9.3 shows a sketch of a set of equally spaced planes that are orthogonal to the plane stretched between k_i and k_s and to $\Delta\bar{k}$. The planes are denoted virtual interference planes. The spacing between the planes is given by

$$l_o = \frac{2\pi}{|\Delta k|} = \frac{\lambda}{2\sin\theta/2} \qquad (9.1.9)$$

where λ is the wavelength of the incident light. If the length of free flight l_f of the molecules is much longer than the length between the virtual interference planes, i.e.,

$$l_f \gg l_o = \frac{2\pi}{|\Delta k|}, \qquad (9.1.10)$$

we can consider the molecules as moving in a kinetic gas, and the region given by eq. 9.1.10 can be defined as the kinetic region. In the opposite case, where the length of the free flight l_f is much smaller than the length between the virtual interference planes, i.e.,

$$l_f = \ll l_o = \frac{2\pi}{|\Delta k|}, \qquad (9.1.11)$$

we look at areas in space and times that are large compared with the microscopic quantities l_f and τ_1, i.e. we can use a macroscopic approach to describe the collective movement of the particles. The region in space defined by eq. 9.1.11 is denoted the hydrodyna nical region for the reason that the movement of particles can be described by a set of hydrodynamical equations.

9.2. The Kinetic Region

Here an attempt is made to describe the conditions in the kinetic region in a case where the interaction between the molecules is weak, so that the contribution from the pair-conditional probability function can be neglected. Employing this assumption we can express the second-order correlation function of the scattered field in the direction of $\bar{k}_s$ by using eqs. 9.1.4 and 9.1.8

$$R_{k_s}(\tau) = I_o \left| \frac{k^2 \alpha(\omega_o) \sin\psi}{4\pi\varepsilon_o r} \right|^2 \frac{N}{V} \int F_s(r,\tau)\, e^{-i\Delta k \cdot r}\, d^3r \qquad (9.2.1)$$

To find the self-conditional probability function, we can use the same technique as employed in chapter 8 in the case of Brownian motion by substituting the relaxation time $1/\beta$ with the time of free flight τ_f. From eqs. 8.2.4 and 8.2.9 we then obtain

$$F_s(r,\tau) = (2/3\ \pi \langle (\Delta\tau)^2 \rangle)^{-3/2}\ e^{-\frac{3r^2}{2\langle (\Delta r)^2 \rangle}} \qquad (9.2.2)$$

where

$$\langle (\Delta r)^2 \rangle = 2\langle v^2 \rangle\, \tau_f^2 \left(\tau/\tau_f - 1 + e^{-\tau/\tau_f} \right) \qquad (9.2.3)$$

where $\langle v^2 \rangle$ is the mean square velocity of the molecules.

By Fourier transforming F_s we obtain

$$R_{k_s}(\tau) = I_o \left| \frac{k^2 \alpha(\omega_o) \sin\psi}{4\pi\varepsilon_o r} \right|^2 \frac{N}{V} e^{-\frac{1}{6}\langle (\Delta r)^2 \rangle (\Delta k)^2} \qquad (9.2.4)$$

The mean free path l_f is related to the time of free flight by

$$l_f = \left(\tfrac{1}{3}\ v^2 \right)^{1/2} \tau_f .$$

The argument in the exponential function in eq. 9.2.4 can then be written as:

$$\frac{1}{6}\langle(\Delta r)^2\rangle(\Delta k)^2 = (l_f \Delta k)^2 (\tau/\tau_f - 1 + e^{-\tau/\tau_f}) . \qquad (9.2.5)$$

The definition of the kinetic region implies that $l_f \Delta k \gg 2\pi$, which means that only the short-time behaviour of eq. 9.2.5 is considered, i.e.

$$\frac{1}{6}\langle(\Delta r)^2\rangle(\Delta k)^2 \approx \frac{1}{6}\langle v^2\rangle \tau^2 . \qquad (9.2.6)$$

Insertion of eq. 9.2.5 into 9.2.4 yields for the second-order correlation function

$$R_{k_s}(\tau) = I_o \left| \frac{k^2 a(\omega_o) \sin\psi}{4\pi\varepsilon_o r} \right|^2 \frac{N}{V} e^{-\frac{1}{6}\langle v^2\rangle \tau^2} . \qquad (9.2.7)$$

The spectral density of the scattered field becomes Gaussian, as seen from eq. 9.2.7.

The above calculation shows that the self-conditional probability function gives rise to a Lorentzian spectral density when passing from the kinetic into the hydrodynamic region.

9.3. The Hydrodynamic Region

In a hydrodynamic description we deal with macroscopic quantities that can be deduced from the related microscopic quantities by a suitable averaging of these quantities over a macroscopic time and length scale. This means that a hydrodynamic description is an asymptotical approach with a validity in the range of long wavelengths and small frequencies. In a hydrodynamic approach the transport phenomena are described by a set of transport equations[45] which is essentially a set of conservation laws for the number density, momentum density and energy density. The set of transport equations is incomplete, and in order to solve the equations, i.e. in respect to our subject to find the density-density correlation function, we must specify some thermodynamic conditions governing the system. Under the condition of local thermodynamic equilibrium in the medium, the set of equations can be solved, and the Landau-Placzek formula is obtained for the density-density correlation function. The calculation giving the Landau-Placzek formula is rather comprehensive and is omitted here. The result is given, and for the details of the calculation reference is made to a paper of Kadanoff and Martin[46].

Another approach in the hydrodynamic region is a relaxation theory of Mandel'shtam and Leontovich[47], which tends to describe the circumstances

at high frequencies and small wavelengths, where the Landau-Placzek formula is not valid.

The main feature of light scattering in the hydrodynamic region can be found from simple phenomenological considerations. By assuming local thermodynamic equilibrium, we can express the fluctuation point of the number density by means of the fluctuations in the local pressure and temperature.

$$\Delta n(r,t) = \left(\frac{\delta n}{\delta p}\right)_T \Delta p(r,t) + \left(\frac{\delta n}{\delta T}\right)_p \Delta T(r,t). \tag{9.3.1}$$

As known, pressure fluctuations in a gas or a liquid give rise to sound waves in a medium. The sound waves are propagated with the velocity of sound and are damped due to the frictional forces in the medium. In a mode description the Fourier-transform of the pressure fluctuation in space must then satisfy the differential equation of a damped harmonic oscillator, i.e. with

$$\Delta p_k(t) = \int \Delta p(\bar{r},t)\ e^{-i\bar{k}\cdot\bar{r}}\ d^3r$$

we obtain:

$$\Delta\ddot{p}_k + 2\alpha_k \Delta\dot{p}_k + \Omega_k^2 \Delta p_k = 0\ , \tag{9.3.2}$$

where α_k is a damping term related to the viscosities of the medium, and ω_k the frequencies of the k^{th} mode. The velocity of sound is the group velocity and it is given by the relation

$$c_s = \frac{\delta\omega_k}{\delta k}\ . \tag{9.3.4}$$

The dispersion relation for Ω_k in gases and liquids can be written as

$$\omega_k = kc_s(k)\ . \tag{9.3.5}$$

From eq. 9.3.2 we find the correlation function for the pressure fluctuations

$$\langle \Delta p_k(t)\ \Delta p_k^*(t+\sigma)\rangle = \langle \Delta p_k(o)\ \Delta p_k^*(o)\rangle\ e^{-\alpha_k|\tau|}\ \cos\left(\sqrt{\Omega_k^2-\alpha_k^2}\,\tau\right) \tag{9.3.6}$$

For the temperature fluctuation, the equation of thermal conductivity yields

$$\Delta T(r,t) - \varkappa \nabla^2 \Delta T(r,t) = 0 \tag{9.3.7}$$

where $\varkappa$ is the thermal conductivity. By Fourier-transforming eq. 9.3.7 in space we obtain:

$$\Delta T_k + \varkappa k^2 \Delta T_k = 0 \,. \tag{9.3.8}$$

From eq. 9.3.8 we obtain the correlation function for the temperature fluctuation:

$$\langle \Delta T_k(t)\ \Delta T_k^*(t+\tau) \rangle = \langle \Delta T_k(o)\ \Delta T_k^*(o) \rangle\ e^{-\kappa k^2 |\tau|} \,. \tag{9.3.9}$$

Now, by assuming that the pressure and thermal fluctuations are uncorrelated, we obtain for the density correlation function

$$\begin{aligned} \langle \rho_k(t)\ \rho_k(t+\tau) \rangle = {} & \left(\frac{\delta n}{\delta p}\right)_T^2 \langle |\Delta p_k(o)|^2 \rangle\ e^{-\alpha_k |\tau|}\ \cos\left(\sqrt{\Omega_k^2 - \alpha_k^2}\ \tau\right) \\ & + \left(\frac{\delta n}{\delta T}\right)_p^2 \langle |\Delta T_k(o)|^2 \rangle\ e^{\varkappa k^2 |\tau|} \end{aligned} \tag{9.3.10}$$

Together with eq. 9.1.4, the above equation implies that the spectral density of the scattered light is composed of three Lorentzians: one unshifted, the Rayleigh peak, due to the thermal fluctuations in the medium, and two shifted, the Mandel'shtam-Brillouin doublets, due to the pressure fluctuation in the medium. The frequency shift is given by

$$\Delta\omega_{\Delta k} = \sqrt{\Omega_{\Delta k}^2 - \alpha_{\Delta k}^2} \tag{9.3.11}$$

which, for negligible damping, can be written

$$\Delta\omega_{\Delta k} = \Omega_{\Delta k} = c_s \Delta k = \frac{4\pi}{\lambda} c_s \sin\theta/2 \,. \tag{9.3.12}$$

The last equation can be interpreted as a Doppler shift. In the Fourier space pressure fluctuations can be considered as collective excitations, phonons, which scatter the photons from the incoming light beam.

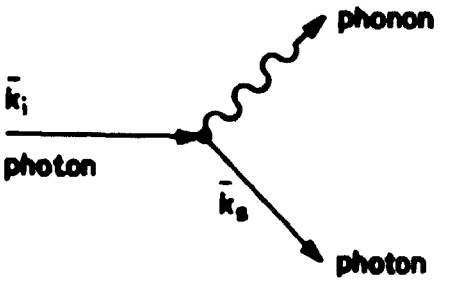


Fig. 9.4

The interaction scheme is sketched in fig. 9.4. By demanding energy and momentum conservation when a phonon is created we obtain:

$$\hbar\bar{k}_i = \hbar\bar{k}_s + \hbar\bar{K} \tag{9.3.13}$$

and

$$\hbar\omega_i = \hbar\omega_s + \hbar\Omega(K) \tag{9.3.14}$$

where $\bar{K}$ is the phonon wavevector.

By solving eqs. 9.3.13 and 9.3.14, we obtain

$$\begin{aligned}\Delta\omega &= \omega_i - \omega_s = \Omega(\Delta\bar{k}) \\ &= \Delta k \cdot \nabla_{\Delta k}\, \Omega(\Delta k) = \Delta k c_s\end{aligned} \tag{9.3.15}$$

i. e. the two Brillouin peaks in the spectral density of the scattered light can be interpreted as a Doppler shift resulting from the creation and annihilation of phonons. The width of the peaks due to the damping term in eq. 9.3.4 can be interpreted as a finite lifetime of the phonon in the scattering medium.

Now, after these preliminary investigations, we write the Landau-Placzek formula without remarks; the interpretation is like that in the previous section. The spectral density of the scattered light becomes:

$$S(\Delta k,\omega-\omega_o) = I_o\left|\frac{k_o^2\alpha(\omega_o)\sin\psi}{4\pi\varepsilon_o r}\right|^2 \frac{\langle|\rho_{\Delta k}(o)|^2\rangle}{k_B T}$$

$$\times (\Delta k)^2\left[\frac{(\Delta k)^2 c_s^2 \Gamma \frac{c_v}{c_p} - D_T(1-c_v/c_p)((\omega-\omega_o)^2 - (\Delta k)^2 c_s^2)}{((\omega-\omega_o)^2 - (\Delta k c_s)^2)^2 + ((\Delta k)^2\Gamma(\omega-\omega_o))^2}\right.$$

$$\left. + \frac{D_T(1-c_v/c_p)}{(\omega-\omega_o)^2 + ((\Delta k)^2 D_T)^2}\right] \qquad (9.3.16)$$

with

$$D_T = \kappa/c_p \qquad (9.3.17)$$

$$\Gamma = D_\ell + D_T(c_p/c_v - 1)$$

and $$D_\ell = \frac{\xi + 4/3\eta}{n_o m}$$

where c_p and c_v denote the heat capacities at constant pressure and volume, κ is the thermal conductivity, η the viscosity and ξ the bulk viscosity. n_o denotes the number density and m the mass of the molecules.

From eq. 9.3.16 one finds that the Brillouin shift is given by

$$\omega_B = \Delta k c_s \sqrt{1 - \frac{(\Delta k)^2\Gamma^2}{2c_s^2}}\,. \qquad (9.3.18)$$

In the cases where the damping effect is negligible, eq. 9.3.18 becomes

$$\omega_B = \Delta k c_s\,. \qquad (9.3.19)$$

The width of the Brillouin peak is

$$\Delta\omega_B = 2\omega_B\left[\sqrt{1 + \frac{(\Delta k)^2\Gamma}{\omega_B^2}((\Delta k)^2 c_s^2 - \tfrac{1}{4}(\Delta k)^4\Gamma^2)^{1/2}} - 1\right] \qquad (9.3.20)$$

which, in the case of negligible damping, reduces to

$$\Delta\omega_B \approx (\Delta k)^2\Gamma. \qquad (9.3.21)$$

As stated earlier, the validity of the Landau-Placzek formula is limited to long wavelengths and small frequencies. Using eq. 9.3.18 we can express this fact more explicitly. The condition for the validity of eq. 9.3.16 is then

$$\frac{(\Delta k)^2 \Gamma^2}{2c_s^2} << 1 \qquad (9.3.22a)$$

or

$$\omega_B << \sqrt{2}\, c_s^2/\Gamma . \qquad (9.3.22b)$$

In the case of some organic liquids such as carbon tetrachloride and benzene that have a large bulk viscosity, the right-hand term in eq. 9.3.22b becomes of the order of 15 Ghz and, as the Brillouin shift is in the order of 5 Ghz, the condition can hardly be said to be fulfilled. The width of the Brillouin peak should further be in the order of 2.5 Ghz, which implies that no peak should be observed in the actual experiment. In the experiment that we performed, the Brillouin peak was clearly visible and the width was about a magnitude smaller than that calculated. In the relaxation theory of Mandel'shtam and Leontovich the damping term (see eq. 9.3.2) due to the bulk viscosity is related to the Brillouin shift by the relation

$$\alpha_{\Delta k} = \frac{c_s}{2c_s^o} \left(\left(\frac{c_s^\infty}{c_s^o}\right)^2 - 1\right) \frac{\omega_B^2 \tau}{1+\omega_B^2 \tau^2} \qquad (9.3.23)$$

where c_s^o and c_s^∞ are the velocities of sound at zero and infinite frequency, respectively. τ is a relaxation time.

From eq. 9.3.20 we see that $\alpha_{\Delta k}$ varies as ω_B squared for small frequencies, as predicted in the Landau-Placzek formula, whereas the $\alpha_{\Delta k}$ becomes constant for larger frequency. The velocity of sound is related to the Brillouin shift by the relation:

$$\frac{c_s^o}{c_s} = 1 - \frac{1}{2} \frac{\omega_B^2 \tau^2 \left(\left(\frac{c_s^\infty}{c_s^o}\right)^2 - 1\right)}{1 + \omega_B^2 \tau^2} . \qquad (9.3.24)$$

The damping term in the range of small and large frequencies can be obtained from eqs. 9.3.23 and 9.3.24:

$$\alpha^{o}_{\Delta k} = \frac{1}{2}\,\omega_B^2\tau\left(\left(\frac{c_s^{\infty}}{c_s^{o}}\right)^2 - 1\right) \tag{9.3.25}$$

and

$$\alpha^{\infty}_{\Delta k} = \frac{1}{2}\,\frac{1}{\tau}\,\frac{\left(\left(\frac{c_s^{\infty}}{c_s^{o}}\right)^2 - 1\right)}{\left(3 - \left(\frac{c_s^{\infty}}{c_s^{o}}\right)^2\right)}\,. \tag{9.3.26}$$

Now, from eqs. 9.3.21, 9.3.25 and 9.3.26, we find

$$\delta\omega_B = 2\alpha^{\infty}_{\Delta k} = \frac{4}{\Gamma}\,c_s^{o}c_s(1 - c_s^{o}/c_s)^2, \tag{9.3.27}$$

which implies that the damping is inversely proportional to the viscosity in the range of large frequencies, whereas the opposite behaviour can be expected from the Landau-Placzek formula.

A further discussion of the Mandel'shtam-Leontovich theory is given in a book by Fabelinskii[47].

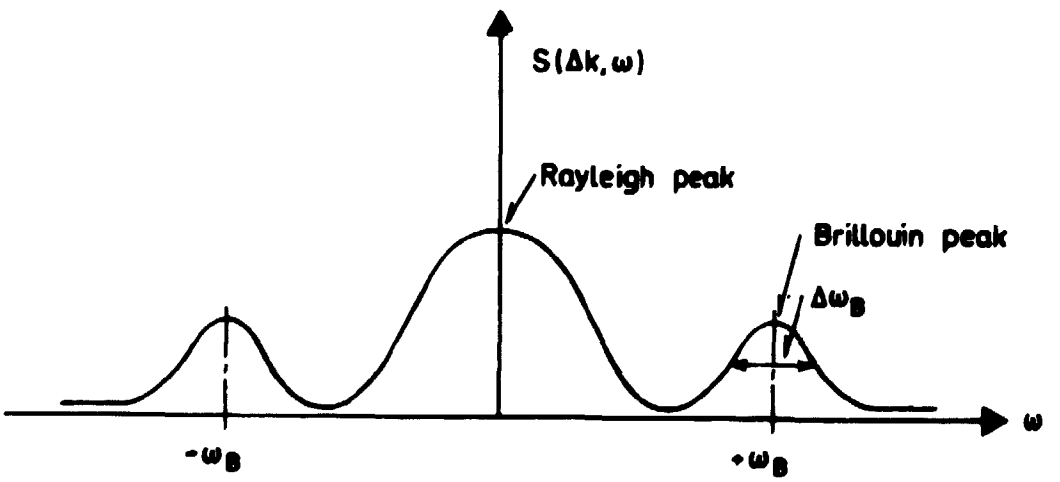


Fig. 9.5

9.4. Experimental Set-Ups

The experimental set-ups are sketched in fig. 9.6. From a 2W Argon laser operating in a single mode, the laser beam is sent through a rectangular sample cell. The scattered light is filtered so that it only transmits some selected direction of the scattered light. Two different types of optical filter were used, one for scattering angles from about 10^{o} to 90^{o}, and the other for a scattering angle of 90^{o}. The first filter (see fig. 9.7) consists of two pin holes separated a distance L, which operates so that only one direction of the scattered light is transmitted. The relation between the scattering angle θ_o and the angle between the laser beam and the optical axis θ_1 (see fig. 9.7) can be found by the relation

$$\cos \theta_1 = n \cos \theta_o \qquad (9.4.1)$$

where n is the refractive index of the scattering medium.

The second optical filter is shown in fig. 9.8. The cylindrical lens together with the following optics operate ideally so that only light from a line segment is transmitted. When the laser beam passes along this line segment, only light scattered at an angle of 90^{o} will be transmitted by the optical filter. The advantage of this filter is that it only collects light from a region around the laser beam and in this way suppresses the effect of parasitic scattered light. Moreover, it increases the solid angle from which the light is picked up compared with the set-ups in fig. 9.7.

After the optical filter, the light passes a spherical mirror Fabry-Perot interferometer (see fig. 9.6). The Fabry-Perot interferometer works like a frequency filter. Figure 9.9 shows that only frequency components that are spaced ν_o, the free spectral range of the Fabry-Perot interferometer, are transmitted. The ratio between the free spectral range and the width of the transmission curves (fig. 9.9) is denoted the "finesse" of the interferometer, i.e.

$$F = \nu_o / \delta\nu . \qquad (9.4.2)$$

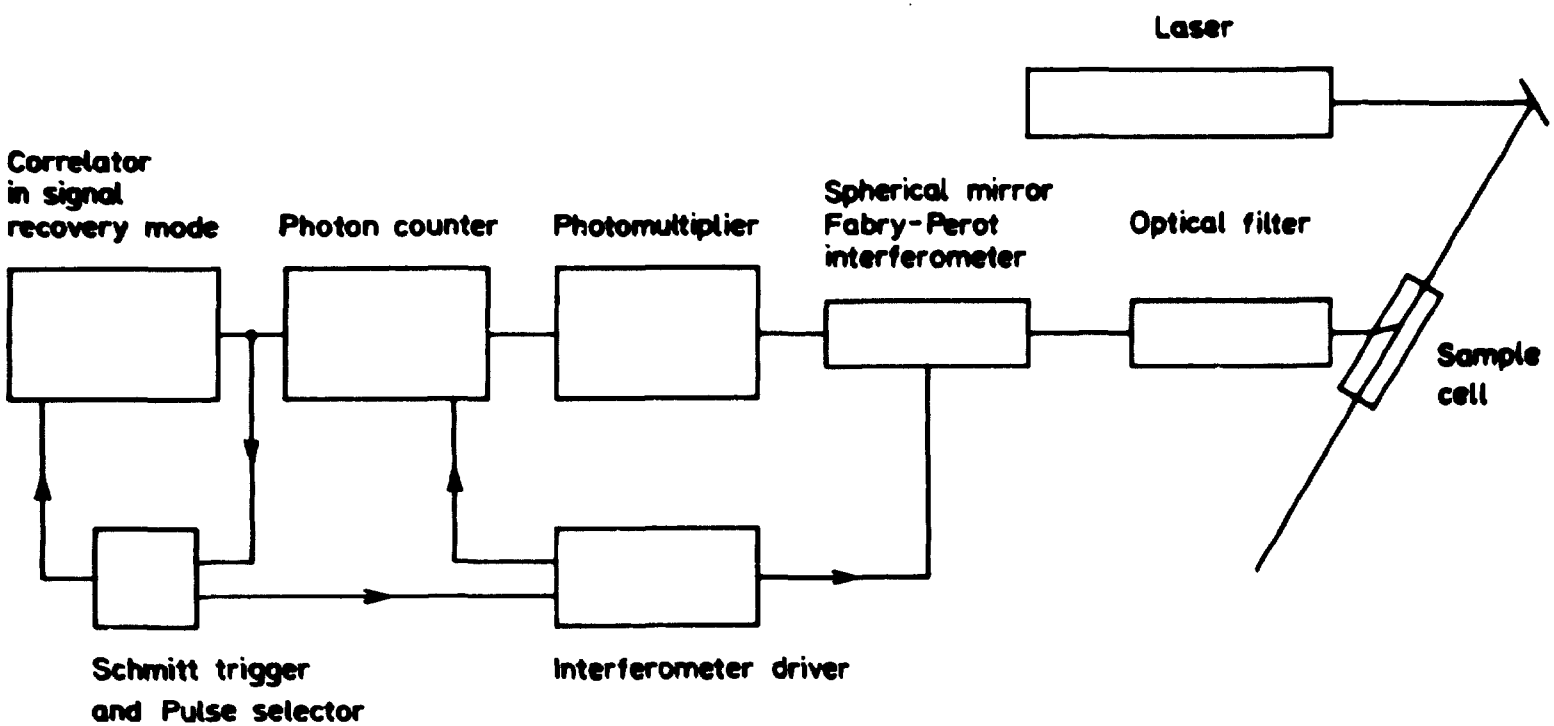

Laser
Correlator
in signal
recovery mode
Photon counter
Photomultiplier
Spherical mirror
Fabry-Perot
interferometer
Optical filter
Sample
cell
Schmitt trigger
and Pulse selector
Interferometer driver

Fig. 9.6

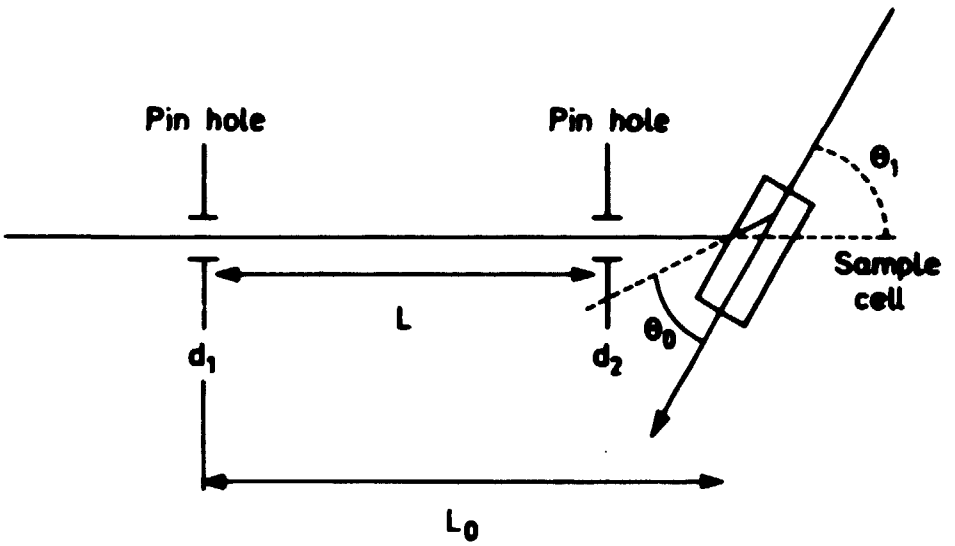

Pin hole
Pin hole
θ_1
Sample cell
L
θ_0
d_1
d_2
L_0

Fig. 9.7

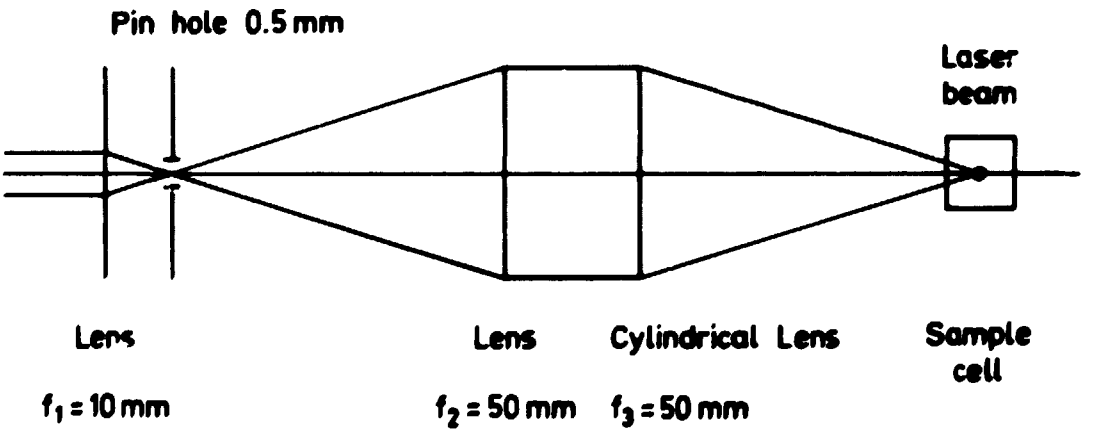

Pin hole 0.5 mm
Laser beam
Lens
Lens
Cylindrical Lens
Sample cell
$f_1 = 10$ mm
$f_2 = 50$ mm
$f_3 = 50$ mm

Fig. 9.8

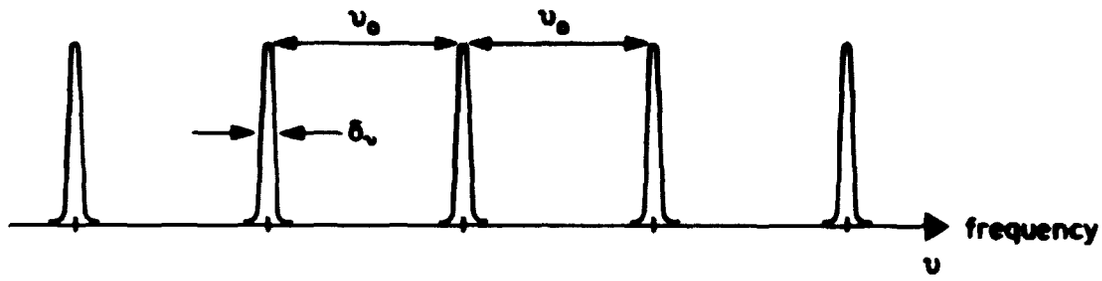


Fig. 9.9

For the Fabry-Perot interferometer used, a finesse can be obtained of about 300 when the system is properly aligned. The transmission frequencies are determined by the spacing between the two spherical mirrors in the interferometer. The mirrors are mounted in a piezo-electric crystal and, by changing the voltage over the crystal, the spacing between the mirrors is changed and thereby the transmission frequency of the interferometer. The voltage across the interferometer is derived from an interferometer driver from where the voltage can be altered in preset steps. The stability of the Fabry-Perot interferometer was measured and it appeared that the transmission frequencies drifted; thus a measurement should be carried out within half a minute to assure stability. The transmitted light from the Fabry-Perot interferometer is detected by a photomultiplier, which is cooled by dry ice in order to suppress the thermally-emitted photo-electrons. A dark current count rate of about one count per second was obtained. The current pulses from the photo-multiplier are counted in a preset counting time by the photon counter. Now, if the Fabry-Perot interferometer was perfectly stable one could obtain the spectrum of scattered light with an arbitrary accuracy by scanning the interferometer across the free spectral range and recording the counts for the different transmission frequencies. The accuracy is determined by the counting time T_0 in the way, that the variance of counts varies as $1/T_0$. The total measuring time T is given by the number of frequency components N it is wished to record and the counting time T_0, i.e.

$$T = NT_0 .$$

In the experiments carried out this time was of the order of one to several minutes, an amount that exceeds the period of stability of the Fabry-Perot interferometer. To avoid stability problems and to obtain reasonable accuracy, we had to average many spectra measured in time intervals that were small compared with the time of stability of the interferometer. This was done by using a HP 3721A correlator in a signal recovery mode. To trigger the correlator, we used the fact that the intensity of the light transmitted at the laser frequency considerably exceeds the intensity level at other frequencies due to the effect of the parasitic scattered light from the walls of the sample cell. Now, when the transmission frequency of the interferometer is equal to the laser frequency, the analogue output of the photon count gives rise to a trigger impulse from the Schmitt trigger that starts the correlator recording the analogue output. When the photon counter has counted for the time T_o, it triggers the interferometer driver so that a new transmission frequency is achieved, and the new analogue output from the photon counter is recorded by the correlator and so on. When the voltage of the interferometer driver reaches a level where the transmission frequency of the interferometer is again equal to the laser frequency, the Schmitt trigger resets the interferometer driver and the measurement restarts, the new spectrum being added to those remaining in the correlator. To select pulses from the Schmitt trigger, a pulse selector was constructed which triggers the correlator at the first pulse and resets the interferometer driver at the second pulse and so on. The above procedure is sketched in fig. 9.10.

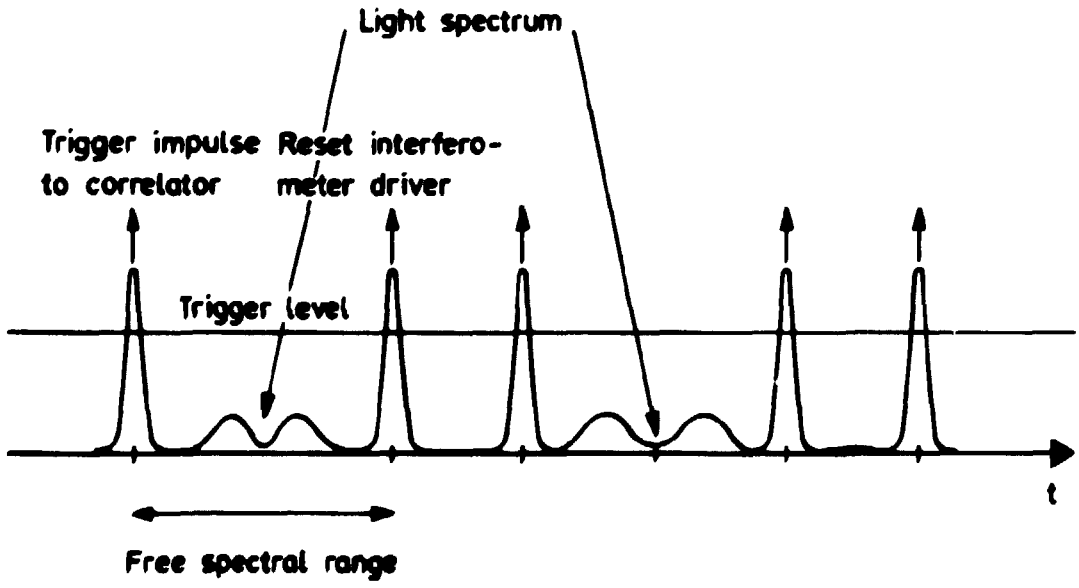


Fig. 9.10

9.5. Spectral Resolution of the System

The spectral resolution of the system is determined by three factors: the spectral resolution of the Fabry-Perot interferometer, the uncertainty of the trigger time, and the resolution of the correlator. If the 100 channels of the correlator cover the free spectral range ν_o of the interferometer, then one channel is eauivalent to

$$\nu_o/100 ,$$

which is the spectral resolution of the correlator. With a properly selected trigger level, the uncertainty of the trigger time was of the order of one to two channels. With a finesse of 300 and a free spectral range of 2Ghz, the spectral resolution of the detecting system can then be estimated to

$$\Delta\nu = \sqrt{\left(\frac{2\nu_o}{100}\right)^2 + \left(\frac{\nu_o}{100}\right)^2 + \left(\frac{1}{F}\right)^2} \approx 45 \text{ Mhz} .$$

9.6. Experimental Results

The results of the experimental investigations are given in tables 9.1 and 9.2. From table 9.2 it is seen that the measured results support the theory of Mandel'shtam and Leontovich, whereas the result calculated from the Landau-Placzek theory is about one magnitude smaller than the values given in (52). The measured values of the velocity of sound of benzene and carbon tetrachloride exceed the values measured for ultrasound[49,50], which also is predicted by the Mandel'shtam-Leontovich theory. Figure 9.12 shows the results from measurements of the Brillouin shift for iso-octane at various angles. The linear behaviour of the frequency shift versus Δk, as predicted by eq.9.3.19. is seen to hold. In fig.9.11 the width of the Brillouin shift is depicted versus Δ^2 for iso-octane. The linear relation predicted by eq.9.3.21 from the Landau-Placzek theory is seen to be reasonable when taking into account that the uncertainty of the measurement of the half-width of the Brillouin peak is about 10%. Figures 9.13a, b, c, 9.14a, b, c and 9.15a, b, c show the measured spectra of the scattered light from various liquids.

The measured values in tables 9.1 and 9.2 and the measured values from refs. 49, 50, 51 and 52 should only be compared with respect to magnitudes for the reason that the temperature and pressure were not controlled during the experiments. The denoted temperature 25°C is only a rough estimate; in the experimental equipment there was no possibility of obtaining a more exact temperature regulation.

9.7. Conclusion

The experimental equipment demonstrated its ability to measure the velocity and the bulk viscosity for different liquids, what is lackning is to perform more exact measurements with control of temperature and pressure. Measurements of gases (CO_2) at atmospheric pressure was carried out without success. For measurements of gases more laser power is needed, and the measurement should be performed at high pressure levels.

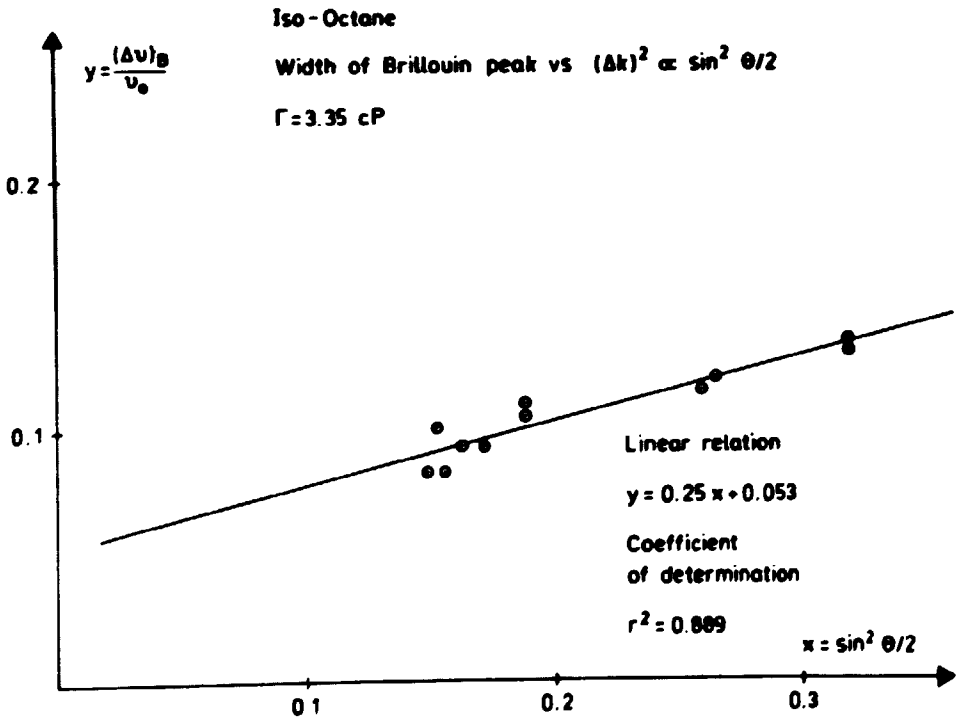


Fig. 9.11

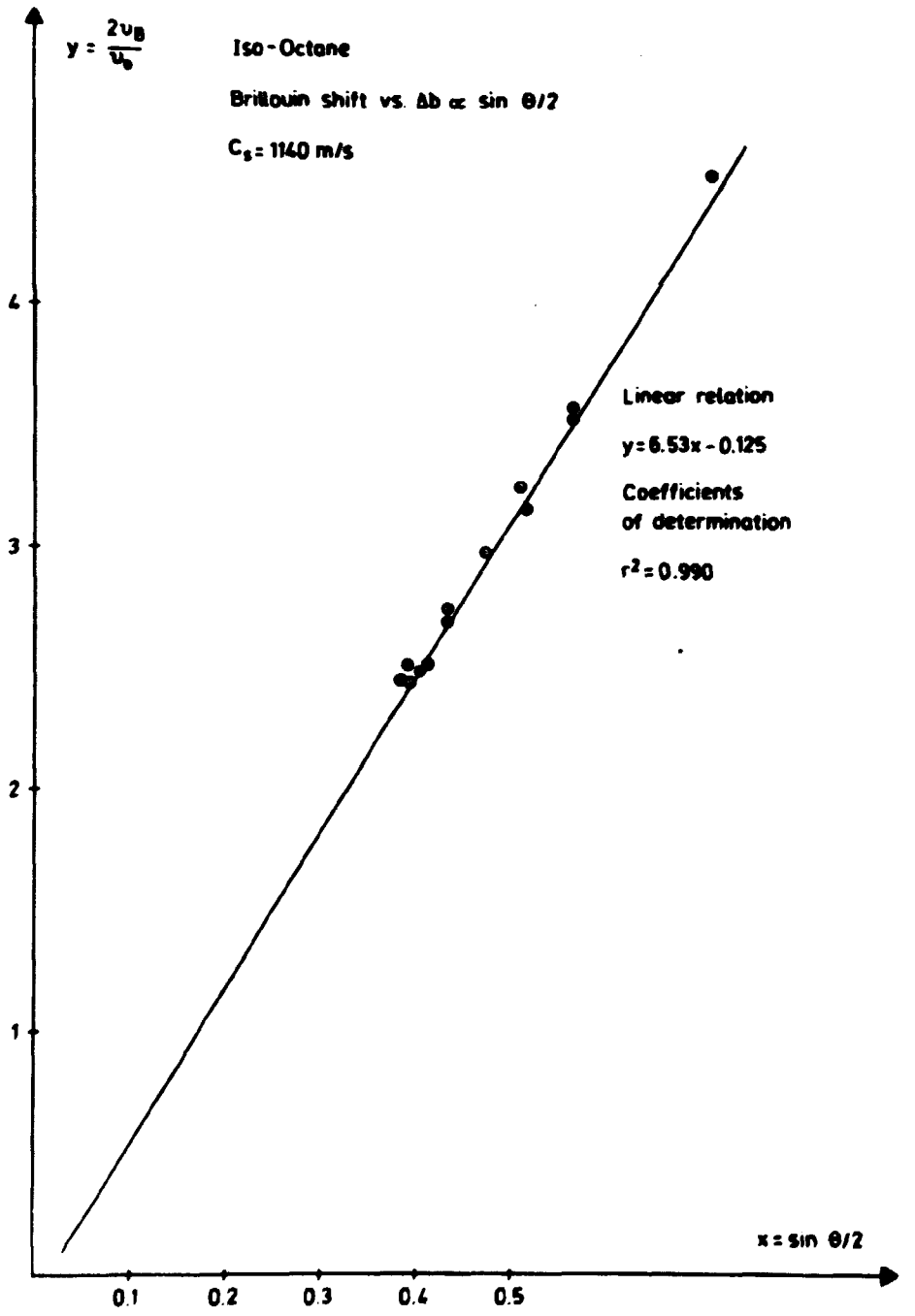
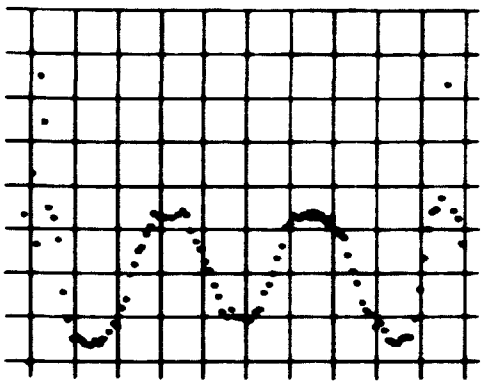
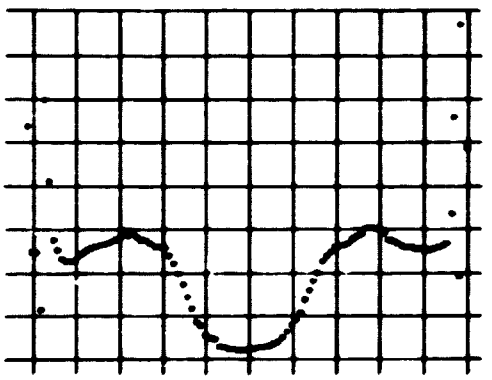

y = 2υB / υ0
Iso-Octane
Brillouin shift vs. Δb ∝ sin θ/2
Cs = 1140 m/s
Linear relation
y = 6.53x - 0.125
Coefficients of determination
r² = 0.990
x = sin θ/2
4
3
2
1
0.1
0.2
0.3
0.4
0.5

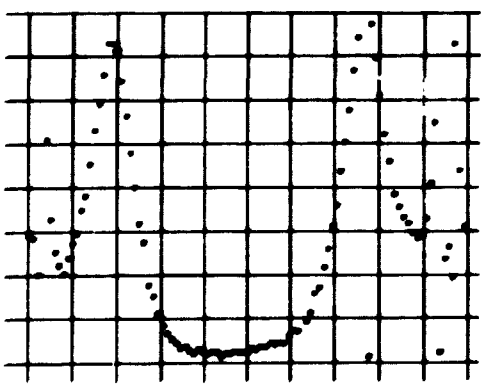
Fig. 9.12



C_6H_6 Benzene
90° scattering angle
Fig. 9.13a

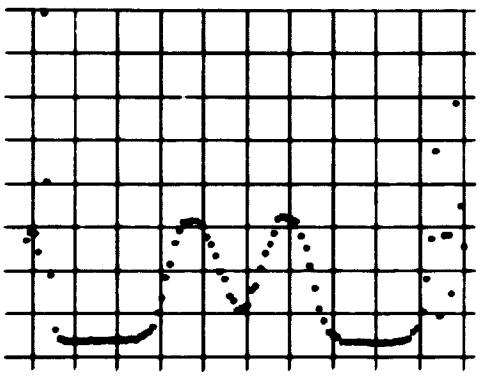


C_8H_{18} Iso-Octane
90° scattering angle
Fig. 9.13b

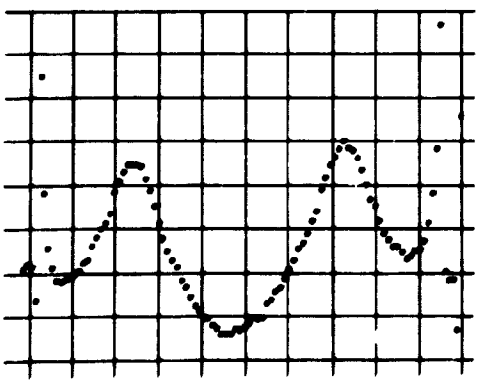


CH_4O Methanol
90° scattering angle
Fig. 9.13c

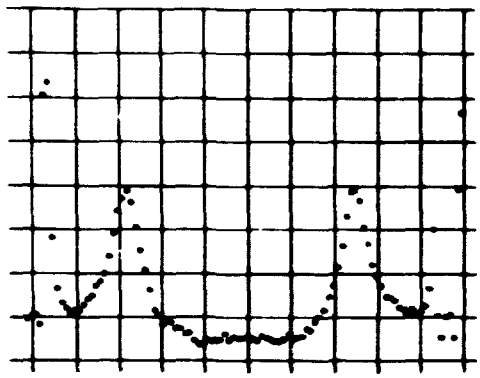
CCl_4 Carbon tetrachloride
90° scattering angle
Fig. 9.14a



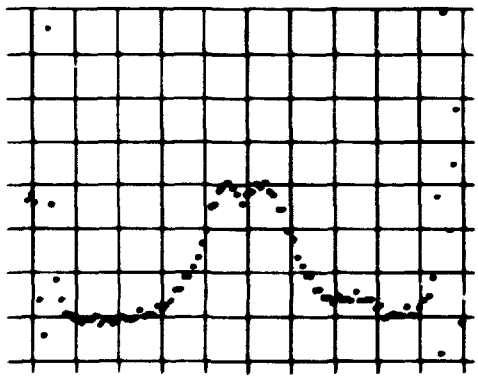
C_3H_6O Acetone
90° scattering angle
Fig. 9.14b



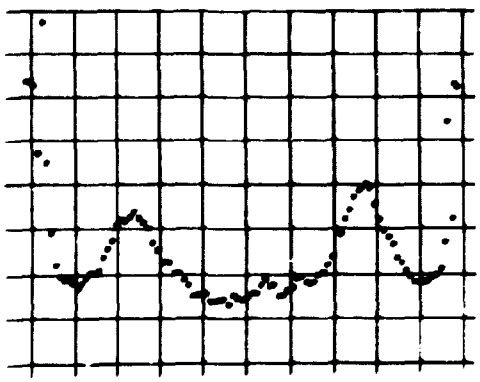
C_6H_{14} N-Hexane
90° scattering angle
Fig 9.14c



C_8H_{18} Iso - Octane
45.5° scattering angle
Fig. 9.15a



C_8H_{18} Iso - Octane
62° scattering angle
Fig. 9.15b



C_8H_{18} Iso - Octane
69° scattering angle
Fig. 9.15c

Table 9.1

		C_8H_{18} Iso-octane	C_6H_{14} n-hexane	C_6H_6 Benzene	C_3H_6O Acetone	CCl_4 Carbon tetra-chloride	CH_4O Methanol
Brillouin shift ν_B GHz	a	4.64	4.51	4.66	4.77	4.65	4.69
Width of the Brillouin peak $\Delta\nu_B$ GHz	a	0.30	0.30	0.35	0.26	0.50	0.28
$\frac{1}{2\pi}\Delta\alpha c_0$ GHz	a	4.48	4.53	4.67	4.78	4.69	4.62
Velocity of sound c_B m/s	a	1106 25°C	1137 25°C	1333 25°C	1215 25°C	1089 25°C	1150 25°C
Velocity of sound m/s	b	1111 20°C	1085 21°C	1295 25°C	1174 25°C	986 25°C	1169 25°C
Velocity of sound m/s	c	-	-	-	-	1084 20°C	-

a) Measured values at t = 25°C and a scattering angle of 90°.
b) Values calculated from refs. (49) and (50).
c) Measured values from ref. (51).

Table 9.2

Bulk viscosity	C_6H_{14} n-hexane	C_6H_6 Benzene	C_3H_6O Acetone	CCl_4 Carbon tetrachloride	CH_4O Methanol
A cP	2.1	2.0	1.6	6.0	1.6
B cP	5.05	77.3	5.05	33.4	5.83
C cP		87		36	

A: Calculated values from the relation $\Delta\omega_B = (qk)^2 F$ due to the London-Placzek formula
B: Calculated values from the Mandel'shtam-Leontovich theory (eqs. 9.3.2.3 and 9.3.2.4
C: Values from ref. (52)

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REFERENCES

1) E. Verdet, Leçons d'Optique Physique I (L'Imprimerie Impériale, Paris, 1869) p. 106.

2) M. Laue, Ann. Phys. 23 (1907) 1.

3) P.H. van Cittert, Physica (The Hague) 1 (1934) 201-210.

4) F. Zernike, Physica (The Hague) 5 (1938) 785-795.

5) L. Mandel and E. Wolf, J. Opt. Soc. Am. 51 (1961) 815-819.

6) R. Hanbury Brown and R.Q. Twiss, Nature (London) 177 (1956) 27-29.

7) E. Wolf. In: Proc. Symp. Optical Masers (John Wiley, New York, 1963) p. 29.

8) R.J. Glauber, Phys. Rev. 130 (1963) 2529-2539.

9) R. Loudon, The Quantum Theory of Light (Clarendon Press, Oxford, 1973) p. 199.

10) J. Perina, Coherence of Light (van Nostrand Reinhold, London, 1972) p. 211.

11) C.L. Mehta, Nuovo Cimento B 45 (1966) 280-282.

12) J.R. Klander and E.C.G. Sudarshan, Fundamentals of Quantum Optics (W.A. Benjamin, New York, 1968) 279 pp.

13) A.E. Siegman, Proc. IEEE 54 (1966) 1350-1356.

14) M. Planck, Verh. Dtsch. Phys. Ges. 2 (1900) 202-204.

15) A. Einstein, Annln. Phys. 17 (1905) p. 132.

16) J.J. Sakurai, Advanced Quantum Mechanics. Chap. 11 (Addison-Wesley, Reading, Mass., 1967).

17) E.A. Power, Introductory Quantum Electrodynamics. Chap. 3 (Longmans, London, 1964) (Mathematical Physics Series, 4) 147 pp.

18) J. Peřina, Coherence of Light (van Nostrand Reinhold, London, 1972) 227-251.

19) P.M. Morse, Thermal Physics (W.A. Benjamin, New York, 1965) 346-359.

20) R.J. Glauber, Phys. Rev. 131 (1963) 2766-2788.

21) E. C. G. Sudershan, Phys. Rev. Lett. 10 (1963) 277-279.

22) A. Einstein, Phys. Z. 18 (1917) p. 121.

23) C. de Witt, A. Blaudin and C. Cohen-Tannoudji, Quantum Optics and Electronics (Gordon and Breach, New York, 1965) p. 144.

24) H. Haken, Z. Phys. 181 (1964) 96-124.

25) P. L. Kelley and W. H. Kleiner, Phys. Rev. A 136 (1964) A316-A334.

26) L. Mandel, Proc. Phys. Soc., London 72 (1958) 1037-1048.

27) G. Bedard, Phys. Lett. 21 (1967) 32-33.

28) S. O. Rice, Bell. Syst. Tech. J. 24 (1945) 47-156.

29) R. S. Lawrence, G. R. Ochs, and S. F. Clifford, Appl. Opt. 11 (1972) 239-243.

30) J. W. Strohbehn, Optical Propagation through the Turbulent Atmosphere. In: Progress in Optics 9. Edited by E. Wolf (North-Holland, Amsterdam, 1971) 75-122.

31) R. L. Fante, Proc. IEEE 63 (1975) 1669-1692.

32) C. Fog, A Photon-Statistical Correlator for LDA Application. Paper presented at the Symposium on the Precision of Flow Measurements by Laser-Based Doppler Methods (LDA-Symp. 1975), Copenhagen, 25-28 August 1975.

33) L. Lading, Appl. Opt. 10 (1971) 1943-1949.

34) L. Lading, Opto-electronics 5 (1973) 175-187.

35) R. Loudon, The Quantum Theory of Light (Clarendon Press, Oxford, 1973) p. 214.

36) P. M. Morse, Thermal Physics (W. A. Benjamin, New York, 1965) p. 155.

37) H. Lamb, Hydrodynamics, 6 ed. (Dover, New York, 1945) 597-606.

38) S. Chandrasekhar, Rev. Mod. Phys. 15 (1943) 1-89.

39) D. W. Schaefer and Bruce J. Berne, Phys. Rev. Lett. 28 (1972) 475-478.

40) H. Z. Cummins, N. Knable, and Y. Yeh, Phys. Rev. Lett. 12 (1964) 150-153.

41) S. B. Dubin, J. H. Lunacek, and G. B. Benedek, Proc. Nat. Acad. Sci. USA 57 (1967) 1164-1171.

43) R. V. Edwards, J. C. Angus, and J. W. Dunning Jr., J. Appl. Phys. 44 (1973) 1694-1698.

44) P. di Porto, B. Crosignani, and M. Bertolotti, J. Appl. Phys. 40 (1969) 5083-5087.

45) N. A. Clark, Inelastic Light Scattering from Thermal Fluctuations in Gases (Dept. of Physics, M. I. T., Cambridge, Mass., May 1970) 38-154.

46) L. P. Kadanoff and P. C. Martin, Ann. Phys. (N. Y.) 24 (1963) 419-469.

47) L. I. Mandel'shtam and M. A. Leontovich, Zh. Eksp. Teor. Fiz 7 (1937) p. 438.

48) I. L. Fabelinskii, Molecular Scattering of Light (Plenum Press, New York, 1968) 622 pp.

49) R. C. Weast (editor), Handbook of Chemistry and Physics. 56. edition. (CRC-Press, Cleveland, Ohio, 1975) Miscellaneous pagination.

50) Landolt-Börnstein, Zahlenwerte und Funktionen aus Naturwissenschaften und Technik. Neue Serie vol. II/5. Molekular-Akoustiks. Von W. Schaffs. (Springer Verlag, Berlin, 1967).

51) Yoshiake Kato and G. A. Zdasiuk, J. Opt. Soc. Am. 65 (1975) 995-998.

52) J. L. Fabelinskii, Adv. Phys. Sci. (USSR) 63 (1957) 474-551.

53) M. Fixman, J. Chem. Phys. 23 (1955) 2074-2079.

54) R. K. Bullough, J. Phys. A 1 (1968) 409-430.

55) L. Rosenfeld, Theory of Electrons (North-Holland, Amsterdam, 1951) 119 pp.

56) C. Kittel, Introduction to Solid State Physics. 4. edition. (Wiley, New York, 1971) p. 381.

APPENDIX A

Properties of Fields with Gaussian Statistics

Let $\bar{V}$ be a complex stochastic vector having a Gaussian distribution with zero mean. The components of the stochastic vector are the electromagnetic fields in different space-time points

$$V = (V(\bar{r}_1 t_1) \ldots\ldots\ldots V(\bar{r}_n \; t_n)). \tag{A.1}$$

The Gaussian distribution function for the complex quantity $\bar{V}$ is denoted

$$P(\bar{V}, \bar{V}^*), \tag{A.2}$$

and the characteristic function for $P(\bar{V}, \bar{V}^*)$ is defined by the relations

$$F(\bar{z}, z^*) = \int P(\bar{V}, \bar{V}^*)\, e^{-iz \cdot \bar{V}^* - iz^* \cdot \bar{V}} = \langle e^{-i\bar{z} \cdot \bar{V}^* - i\bar{z}^* \cdot \bar{V}} \rangle, \tag{A.3}$$

which is the Fourier transformed of $P(\bar{V}, \bar{V}^*)$.

From the characteristic function, we can deduce the moment, i.e. the correlation functions of the fields, by differentiation. From eq. A.3 we obtain:

$$\langle V^*(r_1 t_1)\, V^*(\bar{r}_2, t_2) \ldots V^*(r_p t_p)\, V(\bar{r}_{p+1}, t_{p+1}) \ldots V(\bar{r}_{p+q} t_{p+q}) \rangle$$

$$= (i)^{p+q} \frac{\delta}{\delta z_1} \frac{\delta}{\delta z_2} \cdots \frac{\delta}{\delta z_p} \frac{\delta}{\delta z_{p+1}} F(\bar{z}, \bar{z}^*) \Big|_{z=z^*=0}. \tag{A.4}$$

Now, when $\bar{V}$ has a Gaussian distribution with zero means the characteristic function F becomes

$$F(z, z^*) = e^{-1/2 \langle (\bar{z} \cdot \bar{V}^* + z^* \cdot \bar{V})^2 \rangle}. \tag{A.5}$$

Since the field is only correlated with the complex conjugated field, eq. A.5 can then be reduced to:

$$F(\bar{z}, \bar{z}^*) = e^{-\bar{z}^* \cdot \bar{\bar{U}} \cdot z} \tag{A.6}$$

where

$$\bar{\bar{U}} = \langle \bar{V} \; \bar{V}^* \rangle. \tag{A.7}$$

By reversing eq. A.3, we obtain the distribution $P(V, V^*)$

$$P(\bar{V}, \bar{V}^*) = \frac{1}{2\pi} \int (F(z, z^*)\, e^{i\bar{z}^* \cdot \bar{V}^* + i\bar{z} \cdot \bar{V}^*}\, d\bar{z} d\bar{z}^* = (\det 2\pi \bar{\bar{U}})^{-1}\ e^{-\bar{V}^* \cdot \bar{\bar{U}}^{-1} \cdot \bar{V}} . \tag{A.8}$$

By using eq. A.4, we obtain for the correlation function $\Gamma^{(p,q)}$

$$\begin{aligned} &\Gamma^{(p,q)}(\bar{r}_1 t_1 \ldots r_p t_p, \ldots r_{p+q}\ t_{p+q}) \\ &= \langle V^*(r_1 t_1) \ldots V^*(r_p t_p)\, V(r_{p+1}\ t_{p+1}) \ldots V(r_{p+q} t_{p+q}) \rangle \\ &= \delta_{p,q} \sum_{a} U_{1a_1}\ U_{2a_2} \ldots U_{pa_p} \\ &= \delta_{p,q} \sum_{a} \Gamma^{(1,1)}(r_1\ t_1\ r_{a_1},\ t_{a_1}) \ldots \Gamma^{(1,1)}(r_p\ t_p,\ r_p\ t_{a_p}) \end{aligned} \tag{A.9}$$

where $\sum_{a}$ stands for the sum of all possible permutations of the indices

$p+1,\ p+2,\ \ldots\ 2p$.

Equation A.9 is an extended Siegert relation. In the case $p = 2$ we obtain the usual Siegert relation

$$\begin{aligned} \Gamma^{(2,2)}(r_1 t_1\ r_2 t_2,\ r_3 t_3,\ r_4 t_4) &= \Gamma^{(1,1)}(r_1 t_1, n_3 t_3)\Gamma^{(1,1)}(r_2 t_2, r_4 t_4) \\ &+ \Gamma^{(1,1)}(r_1 t_1 n_4 t_4)\, \Gamma^{(1,1)}(r_2 t_2, n_3 t_4) \ . \end{aligned} \tag{A.4}$$

APPENDIX B

The Electric Field in an Inhomogeneous Dielectric Medium

The Maxwell equation in a non-magnetic medium takes the form:

$$\text{MI:} \quad \frac{1}{\mu}\nabla \times \bar{B} = \epsilon \frac{\delta \bar{E}}{\delta t} + \frac{\delta \bar{P}}{\delta t}$$

$$\text{MII:} \quad \nabla \times \bar{E} = - \frac{\delta \bar{B}}{\delta t}$$

$$\text{MIII:} \quad \nabla \cdot \bar{B} = 0$$

$$\text{MIV:} \quad \nabla \cdot \bar{E} = - 1/\epsilon_o \nabla \cdot \bar{P}$$

where $\bar{P}$ is the electric polarization of the medium.

By introducing the magnetic vector potential $\bar{A}$, and the electric potential V, given by the relation

$$\bar{B} = \nabla \times \bar{A} \quad \text{and} \quad \bar{E} = - \frac{\delta \bar{A}}{\delta t} - \nabla V. \tag{B.1}$$

Insertion of eq. (A.1) into the Maxwell equation gives the two equations

$$\nabla^2 \bar{A} - 1/c^2 \bar{A} = - 1/\mu_o \frac{\delta \bar{P}}{\delta t} \tag{B.2}$$

and

$$\nabla^2 V - 1/c^2 V = - 1/\epsilon_o \nabla \cdot \bar{P} \tag{B.3}$$

with the Lorentz condition

$$\nabla \cdot \bar{A} = - 1/c^2 \frac{\delta V}{\delta t} \tag{B.4}$$

Now, the Lorentz condition is identically fulfilled if there exists a vector field $\bar{\Pi}$, so:

$$\bar{A} = 1/c^2 \frac{\delta \bar{\Pi}}{\delta t} \quad \text{and} \quad V = - \nabla \cdot \bar{\Pi} . \tag{B.5}$$

The vector field $\bar{\Pi}$ is called a Hertz vector, and by insertion of eq. (B.5) into eqs. (B.3) and (B.4), we find that the Hertz potential must fulfil the equation:

$$\nabla^2 \bar{\Pi} - 1/c^2 \frac{\partial^2 \bar{\Pi}}{\partial t^2} = -1/\varepsilon_0 \bar{P} . \qquad (B.6)$$

To find a solution of eq. (B.6), we make a Fourier-transformation in time, and then obtain the Helmholtz-equation

$$\nabla^2 \bar{\Pi} + \omega^2/c^2 \bar{\Pi} = -1/\varepsilon_0 \bar{P} \qquad (B.7)$$

which has the solution:

$$\bar{\Pi}(r,\omega) = \frac{1}{4\pi\varepsilon_0} \int G(r, r', \omega)\, \bar{P}(r', \omega)\, d^3r' + \bar{\Pi}_0(r, \omega) , \qquad (B.8)$$

where G is the Green's function

$$G(r, r', \omega) = \frac{e^{i\,\omega/c\,|r-r'|}}{|r-r'|} \qquad (B.9)$$

and $\bar{\Pi}_0$ is a solution of eq. (B.7) in vacuum.

From eqs. (B.8), (B.1) and (B.5), we then obtain for the electric field:

$$\bar{E}(r, \omega) = (\nabla\nabla + \omega^2/c^2) \frac{1}{4\pi\varepsilon_0} \int G(r, r', \omega)\, \bar{P}(r', \omega)\, d^3r + \bar{E}_0(r, \omega) \qquad (B.10)$$

where E_0 is the externally applied electrical field. The electric polarization in the medium is related to the electric field by the relation

$$\bar{P}(r, \omega) = \varepsilon_0\, \bar{\bar{\chi}}(r, \omega) \cdot \bar{E}(r, \omega) , \qquad (B.11)$$

where $\bar{\bar{\chi}}(r, \omega)$ is the electric susceptibility of the medium, and by insertion of eq. (B.11) into (B.10) we obtain

$$E(r, \omega) = \frac{1}{4\pi} (\vec{\nabla}\vec{\nabla} + \omega^2/c^2 \bar{\bar{1}}) \int_{\omega} G(r, r', \omega) \cdot \bar{\bar{\chi}}(r', \omega) \cdot \bar{E}(r', \omega)\, d^3r' + E_0(r, \omega) \qquad (B.12)$$

Equation (B.12) gives the electric field by an integral equation. In a far field approach, i. e. far from areas where the external field and matter interact, eq. (B.12) can be written:

$$E(r, \omega) \approx E_0(r, \omega) - \frac{e^{ikr}}{4\pi} \int k_s \times (k_s \times (\bar{\bar{\chi}}(r', \omega) \cdot \bar{E}(r', \omega))\, e^{-ik_s \cdot r'}\, d^3r' \qquad (B.13)$$

where $\bar{k}_s = k\,\bar{r}/r$ is the direction of observation.

Finally, we now wish to describe the interaction between the electric field and the medium in greater detail.

The electric polarization P depends on the electric field in the medium due to the interaction between the electric field and the particles in the medium. This interaction can in the first order be described as an induction of an electric dipole at a particle by the electric field. In the first order approach the electric polarization will be the dipole density of the medium and we can write:

$$\bar{P}(r,t) = \sum_i \bar{p}_i(t)\ \delta(r-r_i)\ , \tag{B.14}$$

where $\bar{p}_i(t)$ is the dipole-moment of a particle located at the position $\bar{r}_i$.

The dipole moment p_i is induced by the local electric field E_{loc}, i. e. the external field superposed with the induced fields from the other dipoles in the medium. The local field and the Maxwell field are not, in general, identical quantities. The Maxwell field is a macroscopic field, whereas the local field is essentially a microscopic concept. By using eqs. (B.10) and (B.14), we obtain for the local electric field at the position $\bar{r}_i$:

$$\begin{aligned} E^{(i)}_{loc}(r_i,\omega) &= E_o(r_i,\omega) \\ &+ \frac{1}{4\pi\varepsilon_o}\,(\bar{\nabla}_i\bar{\nabla}_i + \omega^2/c^2) \iint G(r_i,r',\omega) \sum_{j\neq i} p_j(t) \\ &\quad \times\ \delta(r'-r_j)\ e^{-i\omega t}\ d^3r'dt\ . \end{aligned} \tag{B.15}$$

As a second approach we will assume that the dipole moment will be linearly dependent on the local electric field, i. e.

$$p_i(\omega) = \bar{\bar{\alpha}}_i(\omega)\cdot E^{(i)}_{loc}(r_i,\omega) \tag{B.16}$$

where $\bar{\bar{\alpha}}_i$ is the polarizability tensor of the i^{th} particle. In the following, we will assume that $\bar{\bar{\alpha}}$ is independent of the particle number.

By Fourier transforming eqs. (B.14) and (B.13) we obtain

$$\begin{aligned} p_i(t) &= \int \bar{\bar{\alpha}}(t-t')\cdot E^{(i)}_{loc}(r_i,t')\ dt' \\ &= \int \bar{\bar{\alpha}}(t-t')\cdot E_o(r_i,t')\ dt' \\ &\quad + \frac{1}{4\pi\varepsilon_o} \int \bar{\bar{\alpha}}(t-t')\cdot(\bar{\nabla}_i\bar{\nabla}_i - \bar{\bar{1}}\,\frac{1}{c^2}\,\frac{\partial^2}{\partial t^2}) \sum \frac{\bar{p}_j\left(t' + \frac{|r_j-r_i|}{c}\right)}{|r_j-r_i|}\ dt' \end{aligned} \tag{B.17}$$

By solving this equation we can find the electric polarization and thereby the electric field in the medium. An approach to solve this microscopic equation can be found in refs. 53, 54), and 55. Another approach to find the local field can be found by cutting a small sphere A around the local point out of the integration volume Ω in eq. (B.10), and then letting the local field be given by the expression:

$$\mathbf{E}^{(i)}_{loc}(\mathbf{r}_i,\omega) = \mathbf{E}_o(\mathbf{r}_i,\omega) + \frac{1}{4\pi\varepsilon_o}\,(\bar{\nabla}_i\bar{\nabla}_i + \omega^2/c^2\,\bar{\bar{1}})\cdot \int_{\Omega/A} G(\mathbf{r}_i,\mathbf{r}',\omega)\;\bar{P}(\mathbf{r}',\omega)\;d^3r' \tag{B.18}$$

By assuming that there are no polarization charges, i.e.,

$$\nabla \cdot \bar{P} = 0\,,$$

we can write eq. (B.16) as:

$$\mathbf{E}^{(i)}_{loc}(\mathbf{r}_i,\omega) = \mathbf{E}_o(\mathbf{r}_i,\omega) + \frac{1}{4\pi\varepsilon_o}\,\bar{\bar{1}}\;\omega^2/c^2 \int_{\Omega/A} G(\mathbf{r}_i,\mathbf{r}',\omega)\;\bar{P}(\mathbf{r}',\omega)\;d^3r' + \frac{1}{4\pi\varepsilon_o}\int_{\Omega/A} \nabla\cdot(\bar{P}(\mathbf{r}',\omega)\;\nabla G(\mathbf{r}_i,\mathbf{r}',\omega))\,d^3r'$$

and further by using Gauss'es theorem we obtain

$$\begin{aligned}\mathbf{E}^{(i)}_{loc}(\bar{r}_i,\omega) = {} & \mathbf{E}_o(\bar{r}_i,\omega) + \frac{1}{4\pi\varepsilon_o}\,\bar{\bar{1}}\;\omega^2/c^2 \int_{\Omega/A} G(\bar{r}_i,\bar{r}',\omega)\;\bar{P}(\bar{r}',\omega)\;d^3r' \\ & + \frac{1}{4\pi\varepsilon_o}\int_{\delta\Omega} \bar{n}\cdot(\bar{P}(\bar{r}',\omega)\;\nabla G(\bar{r}_i,\bar{r}',\omega)\;dS \\ & + \frac{1}{4\pi\varepsilon_o}\int_{\delta A} \bar{n}\cdot(\bar{P}(\bar{r}',\omega)\;\nabla G(\bar{r}_i,\bar{r}',\omega))\,dS\end{aligned} \tag{B.19}$$

where $\bar{n}$ denotes the normal surface, $\delta\Omega$ and δA the surface of Ω and of the sphere A respectively.

Now, by letting the radius of the sphere A go to zero we obtain:

$$\bar{E}^{(i)}_{loc}(r,\omega) = \bar{E}(r_i,\omega) + \frac{1}{3\varepsilon_o}\,\bar{P}(r_i,\omega)\,, \tag{B.20}$$

where E is the Maxwell field given by eq. (B.10). The local field found by this method can be considered as a macroscopic approach to eq. (B.15), and it will only be valid when the interaction between a dipole and its neighbours is not too "strong", or if the dipoles are placed in a special configuration (cubic configuration, see ref. 56). On the other hand, if the dipole

interaction is "strong", it is doubtful whether the assumption about a linear connection between the dipole moment and the local field will apply. The use of eqs. (B.19) and (B.20) together must then assure some degree of consistence.

In the above calculation we have implicitly assumed that the particles were at rest. This assumption can be removed without complication if the particle velocity is much smaller than the velocity of light, a case that is ordinarily fulfilled. For the time-varying dipole moments we then obtain from eqs. (B.14) and (B.18)

$$\bar{p}_i(t) = \frac{1}{2\pi} \int \bar{\bar{\alpha}}(t-t') \cdot \bar{E}(r_i(t),t')\, dt' + \frac{1}{6\pi\varepsilon_o} \int \bar{\bar{\alpha}}(t-t') \cdot \bar{P}(r_i(t),t')dt' \tag{B.21}$$

and by insertion of eq. (B.19) into (B.12) we obtain

$$\bar{P}(r,t) = \frac{1}{2\pi} n(r,t) \left[\int \bar{\bar{\alpha}}(t-t') \cdot \bar{E}(r,t')\, dt' + \frac{1}{3\varepsilon_o} \int \bar{\bar{\alpha}}(t-t') \cdot \bar{P}(r,t')\, dt' \right] \tag{B.22}$$

where $n(r,t)$ is the particle number density.

By Fourier transforming eq. (B.20) we obtain:

$$P(r,\omega) = \frac{1}{2\pi} \int \bar{\bar{\alpha}}(\omega') \cdot \bar{E}(r,\omega')\, n(r,\omega-\omega')\, d\omega' + \frac{1}{6\pi\varepsilon_o} \int \bar{\bar{\alpha}}(\omega') \cdot \bar{P}(r,\omega')\, n(r,\omega-\omega')\, d\omega'. \tag{B.23}$$

This equation together with eq. (B.10) for the Maxwell field in the medium forms a pair of integral equations, from which the electric field in the medium can be determined.

When the number density is independent of time, eq. (B.21) will have a simple solution:

$$P(r,\omega) = \left(\bar{\bar{1}} - \frac{1}{3\varepsilon_o} n(\bar{r})\, \bar{\bar{\alpha}}(\omega)\right)^{-1} \cdot n(\bar{r})\, \bar{\bar{\alpha}}(\omega) \cdot E(r,\omega) \tag{B.24}$$

which, in the case where $\bar{\bar{\alpha}}$ is a scalar, can be expressed as:

$$P(r,\omega) = (\varepsilon-\varepsilon_o)\, \bar{E} = \frac{n(r)\, \alpha(\omega)}{1-1/3\varepsilon_o\, n(r)\, \alpha(\omega)}\, \bar{E}$$

giving the Clausius-Mosotti relation

$$\frac{\varepsilon-\varepsilon_o}{\varepsilon+2\varepsilon_o} = \frac{1}{3}\, n(r)\, \alpha(\omega)\,. \tag{B.25}$$

Oridinarily $\varepsilon \simeq \varepsilon_o$, meaning that

$$\frac{1}{3}\, \varepsilon_o\, n(r)\, \alpha(\omega) \ll 1\,, \tag{B.26}$$

which further implies that the last term in eqs. (B.22) and (B.24) can be neglected in many applications. If the condition given in eq. (B.26) is not fulfilled, the situation will be more complicated and some saturation phenomena must be expected, as in the case of light beam propagation through strongly turbulent media.

Now, assuming that the condition in eq. (B.26) is fulfilled, we then find the integral equation for the Maxwell field:

$$\begin{aligned} E(r,\omega) = {} & E_o(r,\omega) + \\ & + \frac{1}{8\pi^2\varepsilon_o}\, (\overline{\nabla\nabla} + \overline{\overline{1}}\, \omega^2/c^2) \int G(r,r',\omega)\, \overline{\overline{\alpha}}(\omega') \\ & \qquad E(r',\omega)\, n(r,\omega-\omega')\, d\omega d^3r'\,. \end{aligned} \tag{B.27}$$

In a far field approach, eq. (B.27) becomes:

$$\begin{aligned} E(\bar{r},\omega) \simeq {} & E_o(r,\omega) \\ & - \frac{1}{8\pi^2\varepsilon_o}\, \frac{e^{ikr}}{r}\, \bar{k}_s \times (\bar{k}_s \times (\int \overline{\overline{\alpha}}(\omega') \cdot E(r',\omega') \\ & \qquad n(r,\omega-\omega')\, e^{-i\bar{k}_s\cdot r'}\, d\omega' d^2r')\,. \end{aligned} \tag{B.28}$$

From eq. (B.26) one finds that the scattered field to the first order

$$\begin{aligned} E_s(\bar{r},\omega) = {} & - \frac{1}{8\pi^2\varepsilon_o}\, \frac{e^{ikr}}{r}\, k_s \times (k_s \times \int \overline{\overline{\alpha}}(\omega') \cdot \\ & \qquad \bar{E}_o(r',\omega')\, \Delta n(r',\omega-\omega')\, e^{-i\bar{k}_s\bar{r}'}\, d\omega'\, d^3r') \end{aligned} \tag{B.29}$$

where Δn denotes the fluctuating part of the number density.

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